Calhoun
Institutional Archive of the Naval Postgraduate School

Calhoun: The NPS Institutional Archive

DSpace Repository

Theses and Dissertations 1. Thesis and Dissertation Collection, all items

1953

An investigation of the radionuclides of arsenic produced by cyclotron bombardment of germanium with 15 Mev deutrons

Watters, Harry J.; Fagen, John F.

Massachusetts Institute of Technology

http://hdl.handle.net/10945/14351

Downloaded from NPS Archive: Calhoun

DUDLEY
KNOX
LIBRARY

Calhoun is the Naval Postgraduate School's public access digital repository for research materials and institutional publications created by the NPS community. Calhoun is named for Professor of Mathematics Guy K. Calhoun, NPS's first appointed -- and published -- scholarly author.

Dudley Knox Library / Naval Postgraduate School
411 Dyer Road / 1 University Circle
Monterey, California USA 93943

http://www.nps.edu/library

AN INVESTIGATION OF THE RADIONUCLIDES OF ARSENIC PRODUCED BY CYCLOTRON BOMBARDMENT OF GERMANIUM WITH 15 MEV DEUTERONS

HARRY J. WATTERS
AND
JOHN F. FAGAN, JR.

Thesis
W27

Library
U. S. Naval Postgraduate School
Monterey, California

17-62

AN INVESTIGATION OF THE RADIONUCLIDES OF ARSENIC PRODUCED BY CYCLOTRON BOMBARDMENT OF GERMANIUM

with

15 Mev DEUTERONS

by

HARRY J. WATTERS
Lieutenant Commander, U. S. Navy
B. S. Purdue University
(1949)

and

JOHN F. FAGAN, JR.
Lieutenant, U. S. Navy
B. S., U. S. Naval Academy
(1945)

SUBMITTED IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF
MASTER OF SCIENCE

ABSTRACT

Title: "An Investigation of the Radionuclides of Arsenic
Produced by Cyclotron Bombardment of Germanium
with 15 Mev Deuterons"

Authors: Harry J. Watters, Lieutenant Commander, U.S. Navy
B. S., Purdue University (1949)

and

John F. Fagan, Jr., Lieutenant, U. S. Navy
B. S., U. S. Naval Academy (1945)

Submitted to the Department of Physics on May 25, 1953 in partial fulfillment of the requirements for the degree of Master of Science.

20653

The arsenic produced by a deuteron bombardment of germanium has been studied to determine the nuclides present in the mixture. Identification of isotopes was made by comparing measured values of half life and maximum β energy with the accepted values. Yield values were determined for each isotope present by 4π solid angle counter measurements.

Counting rates were measured for a period of 53 days with 4π and coincidence counters, obtaining half lives which indicated that the nuclidic mixture was made up of As^{71}, As^{72}, As^{73}, As^{74}, and As^{77}. These indications were confirmed by maximum β energy values obtained by absorption measurements and from γ energies found using a γ-ray scintillation spectrometer. Measurements indicated that the 40 hour half life reported for As^{77} is in error by a significant amount, and that no As^{76} was obtained by this bombardment.

The 4π solid angle counter constructed was shown to have an efficiency of very nearly 100 percent for particles which escape the source. This counter has proven to be a very practical laboratory instrument and detailed instructions for its use are included as an appendix.

The following is a tabular summary of the results of the investigation:

Isotope	Method of decay	Energy (Mev)	$T_{1/2}$	Thick target yield* (μc/μamp-hr)
As^{71}	β^+	0.66	48.2 ± 1.2 hrs.	7.6
As^{72}	β^+	3.25	25.8 ± 0.2 hrs.	64.9
	γ	0.85		
As^{73}	β^-	$0.11 > E_{max} > 0.02$	88.9 ± 9.2 days	1.1
As^{74}	β^+	0.92, 1.49	17.89 ± 0.13 days	5.2
	β^-			
As^{76}	Not present in the mixture			
As^{77}	β^-	< 0.7	$>$70 hours	$5 <$ yield < 15**

* The thick target yield values specified apply if the deuteron beam current was exactly 38 μamps and if the arsenic separation efficiency was 100 percent. Yield values quoted are based on β counting only and do not include orbital electron capture.

** Based on ratios of total β to β^+ counting rates.

Thesis Supervisor: Robley D. Evans

Title: Professor of Physics

ACKNOWLEDGMENTS

The authors wish to express sincere thanks to their thesis advisor, Professor Robley D. Evans, for his interest and advice during the course of this work. Grateful acknowledgment is also made to Doctor Gordon L. Brownell, who suggested the problem, for his constant guidance and for the opportunity to utilize the facilities of the Massachusetts General Hospital Research Laboratory.

Thanks are due to all members of the group in the Radioactivity Center for their interest and suggestions. The opportunity for graduate study provided by the Radioactivity Center and its sponsors is greatly appreciated.

TABLE OF CONTENTS

Section I. Introduction 1

A. Importance of the Investigation 1

B. Results of Previous Investigations . . . 4

Section II. Nuclear Properties to be Measured 6

Section III. Experimental Procedure 7

A. Preparation of Radioactive Arsenic . . . 7

B. Schedule of Observations 7

Section IV. Methods Used in Interpretation of Data 12

A. Half Life 12

B. Absolute β Activity 16

C. Maximum β Energies 16

Section V. Results 22

Bibliography 25

Appendix I. The 4π Counter 27

Appendix II. The Scintillation γ-Ray Spectrometer . . 73

Appendix III. The End Window β Counter 75

Appendix IV. The Coincidence Counter 85

LIST OF FIGURES

Figure No.	Title	Page
1	Arsenic γ-ray energy spectrum for 56 hours after bombardment	10
2	Arsenic γ-ray energy spectrum 52 days after bombardment	11
3	Half-life components of coincidence counter data	13
4	Half-life components of 4π counter data	15
5	Maximum range determination from absorption measurements	17
6	β energy determination from absorption measurements (As^{74})	19
7	β energy determination from absorption measurements (As^{71})	20
8	β energy determination from absorption measurements (As^{72})	21

I. INTRODUCTION

A. Importance of the Investigation

The fact that tumors of the brain take up a large amount of trace metal compared with that taken up by normal brain tissue makes it possible to detect and to actually determine the location of tumors in the human brain. Under the supervision of Dr. Gordon L. Brownell, a group at the Research Laboratory of the Massachusetts General Hospital has developed a suitable tracer technique for the diagnosis and preoperative location of brain tumors, using positron emitting isotopes. After intravenous injection of the tracer material, the patient's head is mechanically scanned in two dimensions by two scintillation counters connected in coincidence. Third dimensional location is obtained from the unbalanced single channel counting rates of the separate counters.

Since January 1953 a large number of patients have been examined using this technique. The results are

outstanding. From many cases clinically diagnosed as borderline, the presence or absence of neoplastic brain tissue has been determined by this method. In all cases where surgery was performed, tumor location obtained by this technique has been confirmed. As yet no known incorrect diagnoses have been made. In addition to providing more quantitative information than is available from clinical diagnosis, this method provides the left-to-right localization which is difficult and often impossible to obtain clinically.

Radioactive arsenic was selected as the tracer metal because of several considerations. Arsenic is readily available from a deuteron bombardment of germanium. The half lives of arsenic isotopes fall within an acceptable range for tracer utilization. Most of the γ-rays emitted from arsenic isotopes are soft, thus decreasing harmful biological effects due to radiation. A very important advantage is that a large percentage of arsenic activity consists of positron emission. Precision measurements with very high resolution may be made on the resulting annihilation radiation.

The tracer arsenic is not injected until several days after bombardment. During this period any short-

lived activity present decays to a negligible value compared with that of the 17.5 day isotope(11,13) and does not affect the scanning measurements which require a period of approximately two hours. If the half lives and the relative activity percentages of the short-lived isotopes were accurately known, this waiting period could be decreased or even eliminated with a resultant increase in useful activity obtained from a given bombardment.

The purpose of the present investigation is to determine insofar as possible the methods of decay and associated decay energies, half lives, absolute activities, and isotopic yields of the arsenic obtained by the deuteron bombardment of germanium. In addition to decreasing the delay between bombardment and injection, this information may permit the use of short-lived isotopes as tracers. In effect this also decreases the bombardment time required to obtain a given amount of tracer material. It may be desirable to examine a single patient several times over a period of a few weeks. Accurate knowledge of the short-lived activity present may permit frequent injections of a lesser amount of tracer solution while avoiding harmful effects from the chemical toxicity of carrier arsenic present.

B. Results of Previous Investigations

Prior to Sagane's investigations in 1938(1) very little was known about the radionuclides of arsenic. The principal results of his work on the arsenic produced by a deuteron bombardment of germanium, as modified by others, are tabulated below and include all data reported through 1941.

Isotope	Type radiation	Energy (Mev)	Half life	Reference
As^{71}	β^+		60 hour	2, 4
As^{73}	β^+	0.6	88 min	2, 4
As^{74}	β^+	0.9	16 day	1, 4
As^{76}	β^-	1.1	26.8 hour	3, 5, 6
		1.7		
		2.7		
	γ	1.5		
		2.2		
		3.2		
As^{77}	β^-		55-90 day	1, 2, 4
	γ			
As^{78}	γ	0.27	66 min	7

Very little new information was published for several years but commencing in 1948 results were published which

conflicted with much of the previous data. The following is a tabulation of the most reliable data now available on the radioisotopes of arsenic without regard to their method of activation:

Isotope	Type radiation	Energy (Mev)	Half life	Reference
As^{70}	β^+		52 min	9
As^{71}	β^+ (33 %)	0.6	50-60 hour	10, 11, 12
	K (67 %)	0.162		
As^{72}	β^+	0.27	26 hour	9, 11, 19
		0.67		
		1.84		
		2.5		
		3.34		
	γ	0.702		
		0.835		
As^{73}	K	0.052	76-100 day	11
	no β^+			
As^{74}	β^-	0.69, 1.36	17.5 day	11, 13
	β^+	0.92, 1.53		
	$\beta^-/\beta^+ \sim 1.0$ %			
	γ	0.593		
As^{76}	$\beta^+/\beta^- \leq 0.07$ %			
	γ	0.55, 1.21	27.6 hour	14, 18
As^{77}	β^-	0.679, 0.7	40 hour	15, 16, 20
	no γ			

II. NUCLEAR PROPERTIES TO BE MEASURED

Time and equipment limitations prohibited conducting an investigation which could determine actual decay schemes of the active material. With a desire to extract as much information as possible in the time available, attempts were made to determine the following for each isotope of arsenic obtained from the bombardment:

1. Absolute β activity.
2. Half life.
3. Maximum β energies.

In addition it was desirable to obtain information regarding the γ-energies of the mixture of isotopes and the variation of the spectrum with time.

III. EXPERIMENTAL PROCEDURE

A. Preparation of Radioactive Arsenic

A chip of pure germanium metal 1/32 inch thick with dimensions 3/8 inch by 1 inch was used as a target in the M.I.T. cyclotron. This chip was bombarded with 15 Mev deuterons for a period of 20 minutes with an average beam current of 36 μamps.

After bombardment the germanium metal was oxidized to $GeCl_4$ in an evacuated system using gaseous Cl_2. To this was added HCl, H_2O_2, and arsenic carrier after which the bulk of the $GeCl_4$ was distilled out. The arsenic remaining in the solution as As^{+5} was precipitated as a metal by the addition of ammonium hypophosphite. A detailed description of this separation procedure is contained in reference 21.

B. Schedule of Observations

Continuous observations were made of the disintegration rate of the active material by use of the 4π and coincidence counters. In an attempt to ascertain

whether or not the arsenic contained any positron-emitting isotopes having half lives of the order of 1 hour or less (2,3,17,23,24), coincidence counter measurements were made as follows: each minute during the third hour after bombardment, every 5 minutes during the fourth hour, every 10 minutes during the fifth hour, and every 15 minutes during the sixth hour. Thereafter the maximum interval between measurements was adjusted to approximately 1/10th the value of the half life indicated by a continuous plot of counting rate observations.

Due to the time required for preparation of 4π counter sources and the time involved in making absorption measurements with the end window β counter, observations with these instruments were made hourly from the 6th through the 17th hour after bombardment, and thereafter in accordance with the schedule outlined above.

Using the sodium iodide scintillation spectrometer described in Appendix II an initial scan of the energy spectrum up to 3 Mev was made within three hours after bombardment in order to determine the maximum energy γ-rays emitted from the arsenic. With no detectable γ-energies present greater than 1 Mev, an operating range

was chosen which included all γ-energies up to approximately 1.3 Mev. This energy range was scanned continually for the first 72 hours after bombardment (Fig. 1). The high energy range was scanned at intervals during this period with negative results. An additional energy spectrum was obtained 52 days after bombardment (Fig. 2) and as before, no high energy γ-rays were detectable.

Using the end window β counter, counting rates were measured for absorber thicknesses of from zero to 2526 mg/cm^2. Observations were made in accordance with the time schedule previously given.

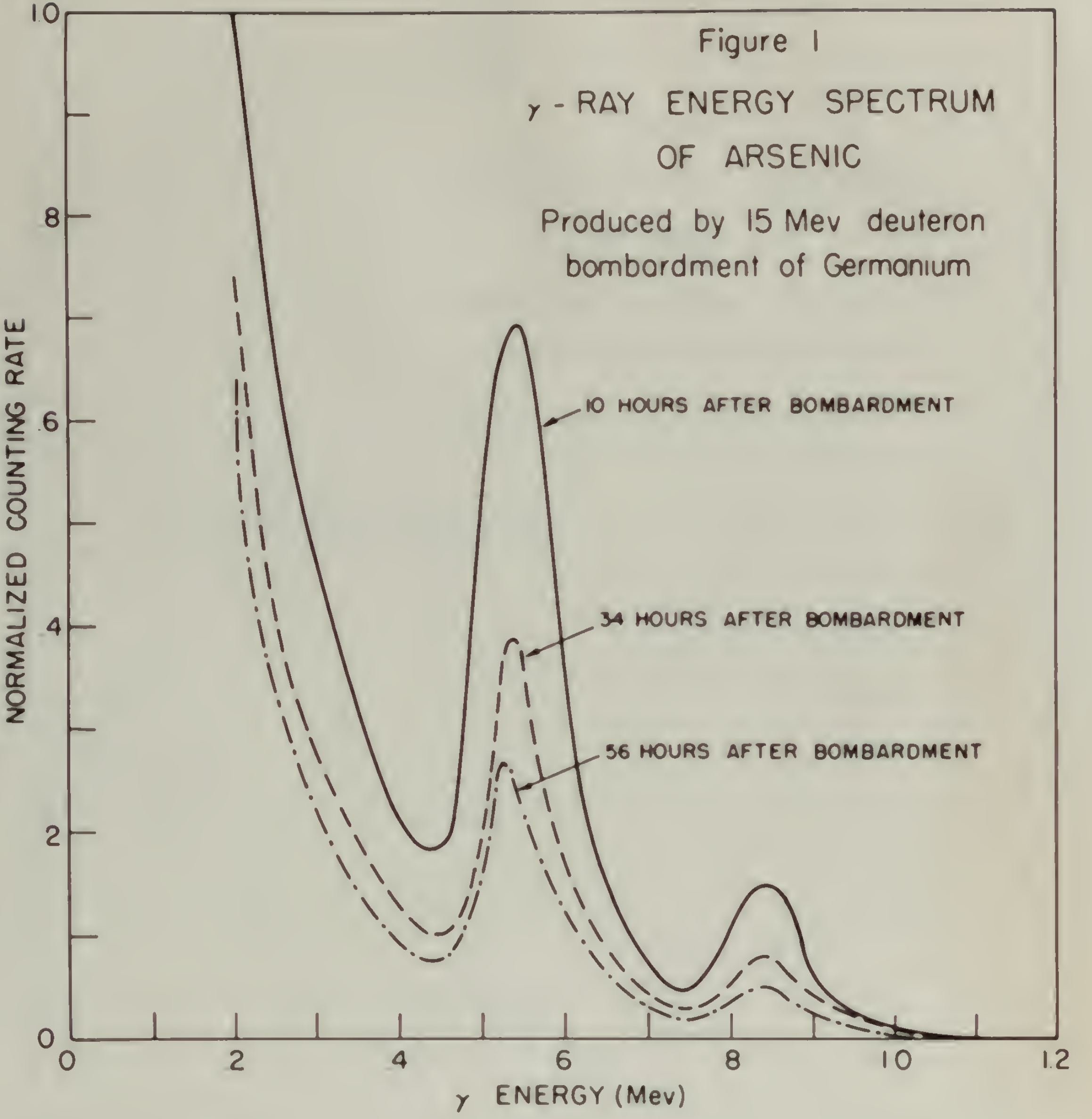


Figure 1

γ - RAY ENERGY SPECTRUM OF ARSENIC

Produced by 15 Mev deuteron bombardment of Germanium

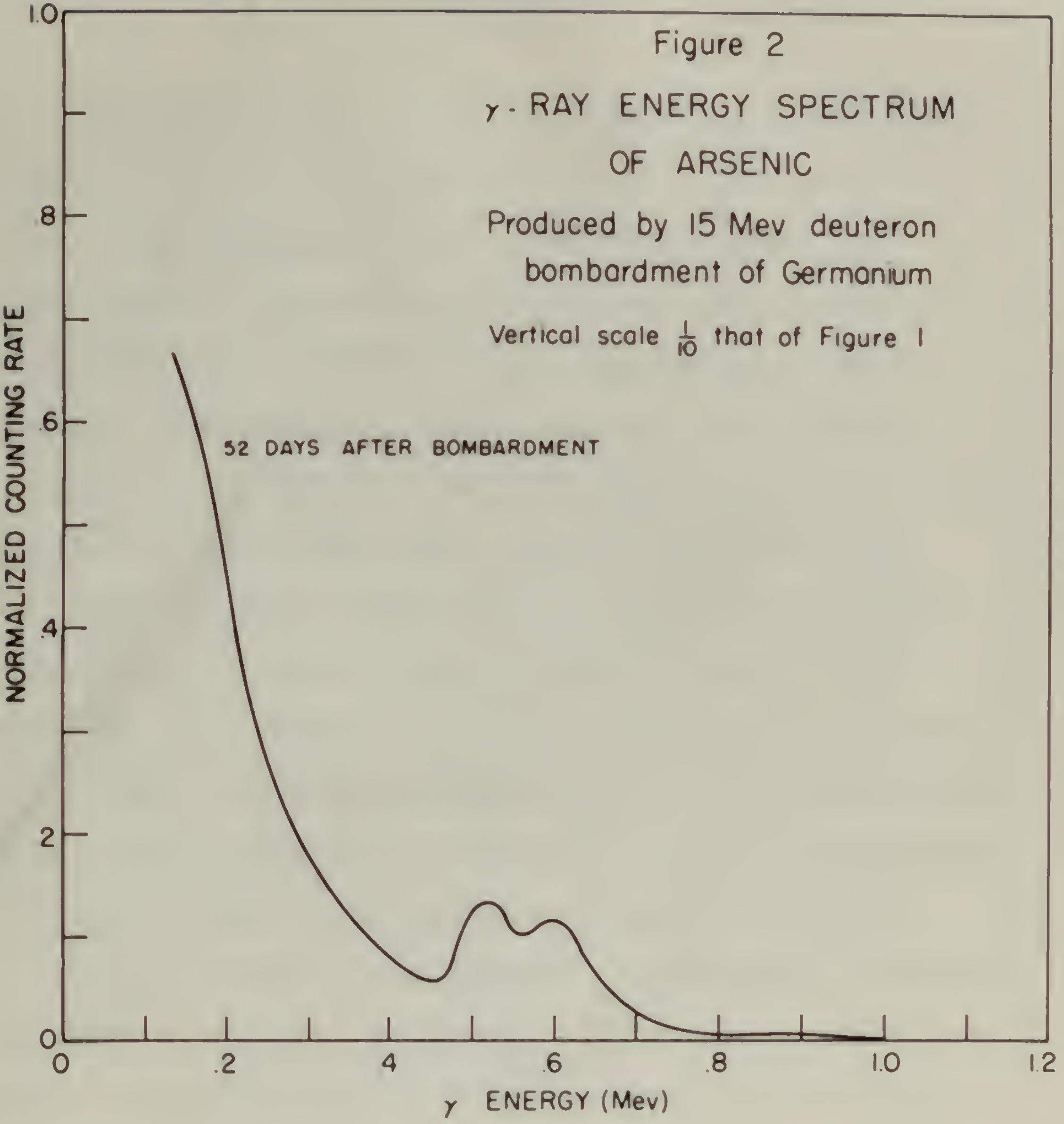


Figure 2

γ - RAY ENERGY SPECTRUM OF ARSENIC

Produced by 15 Mev deuteron bombardment of Germanium

Vertical scale $\frac{1}{10}$ that of Figure 1

IV. METHODS USED IN INTERPRETATION OF DATA

A. Half Life

Observed counting rate, corrected for instrumental error, was plotted on semilog paper as a function of time. Approximately 20 days after bombardment the curve obtained from coincidence measurements assumed a constant slope indicating the presence of a single isotope. Application of the method of least squares to data in the region of constant slope yielded a determination of half life, zero time activity, and their respective standard deviations. Subtraction of values thus obtained from the curve of total counting rate resulted in a residual curve also possessing a constant final slope. Successive application of this method permitted the resolution of 3 straight line components from the data obtained by coincidence counting (Fig. 3).

The 4π counter data included a relatively long-lived component which was not apparent in coincidence measurements. Assuming this to be As^{73} reported as a 0.05 Mev

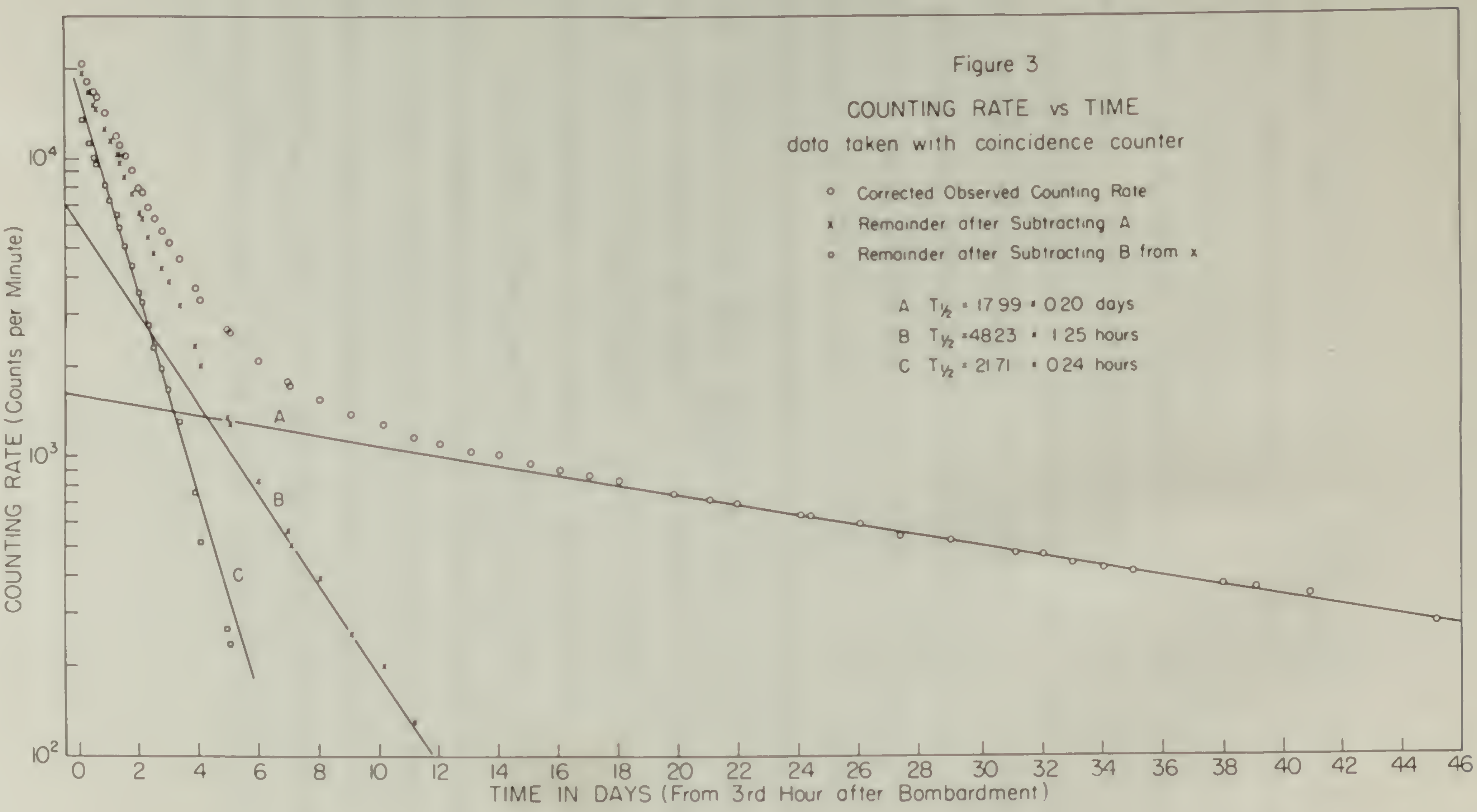


Figure 3

COUNTING RATE vs TIME

data taken with coincidence counter

negatron emitter of half life ~ 80 days,(19) one 4π counter source was covered with 13.7 mg/cm^2 of aluminum foil (a thickness equivalent to ~ 3 times the range of a 0.05 Mev electron) commencing on the 41st day after bombardment. Data obtained with this source plotted as a straight line with a half life of 17.66 days. After subtraction of this 17.66 day activity from the total counting rate curve the constant slope extremity of the residual curve indicated a half life of ~85 days. This procedure permitted early evaluation of data without waiting for the predominance of the 89 day component and the results are in good agreement with the As^{73} method of decay reported by Mei.(19) Successive application of the method of least squares to the 4π counter data resulted in the resolution of 4 straight line components. (Fig. 4)

Half life determinations were also made from semilog plots of counting rate vs time obtained using absorbers of specific thickness with the end window β counter. The method of curve subtraction previously outlined was employed and a total of 3 straight line components were resolved having half lives of 26 hours, 53 hours, and 16.7 days.

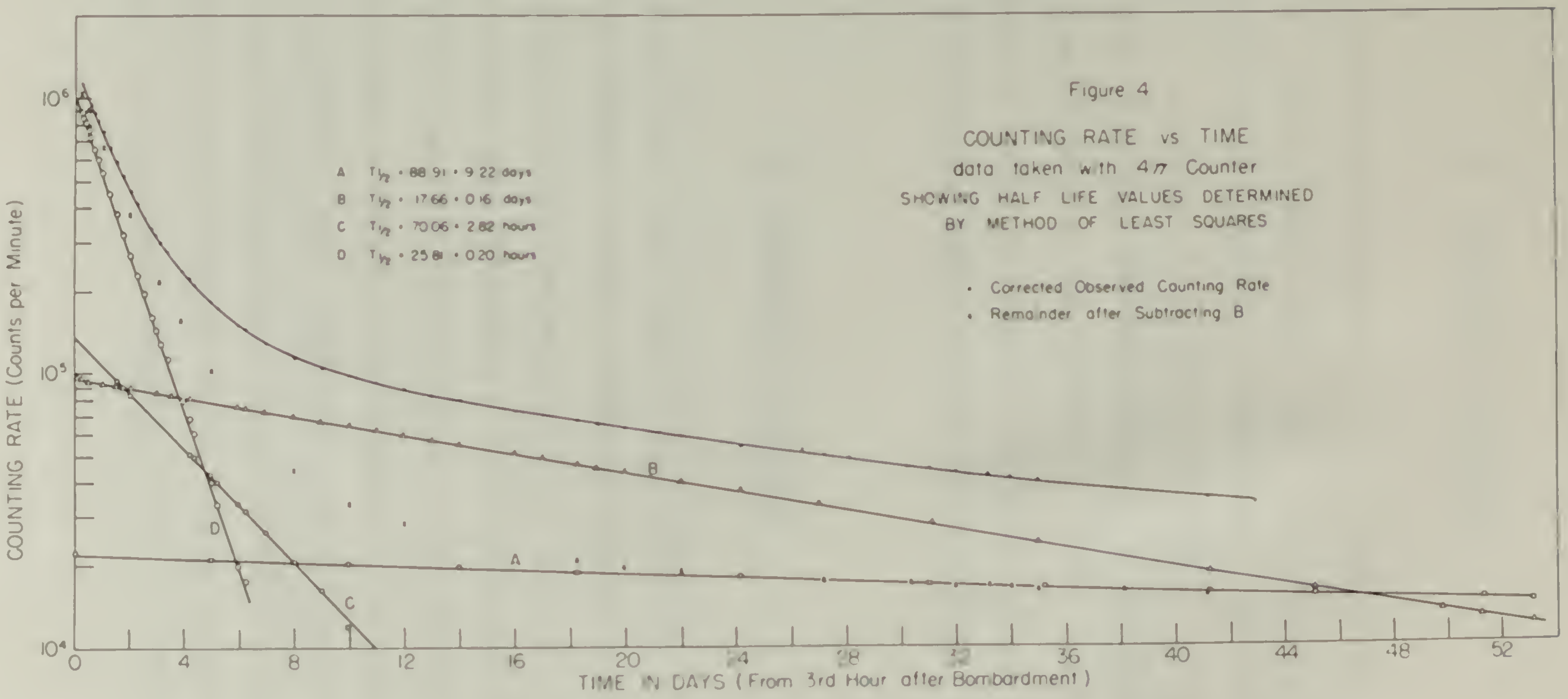


Figure 4

COUNTING RATE vs TIME
data taken with 4π Counter
SHOWING HALF LIFE VALUES DETERMINED
BY METHOD OF LEAST SQUARES

B. Absolute β Activity

Since the efficiency of the 4π counter for β counting is quite high (Appendix I), these data were used in the determination of absolute β activities. The zero time activities obtained in applying the method of least squares to half-life determination were corrected to the time of completion of bombardment. These results can be specified in terms of yield if specific values of deuteron beam current and arsenic separation efficiency are assumed.

C. Maximum β Energies

These values were found from absorption curves obtained by use of the end window β counter (Appendix III). From measurements of maximum range made at various times the energy of the most energetic β was determined for both the 26 hour and the 17.5 day isotopes. The method is illustrated in Fig. 5 which is applicable to the 26 hour isotope.

In addition, mass absorption coefficients were determined from semilog plots of counting rate vs absorber thickness taken at various times. Using these values maximum β energies were determined for the 17.5 day and

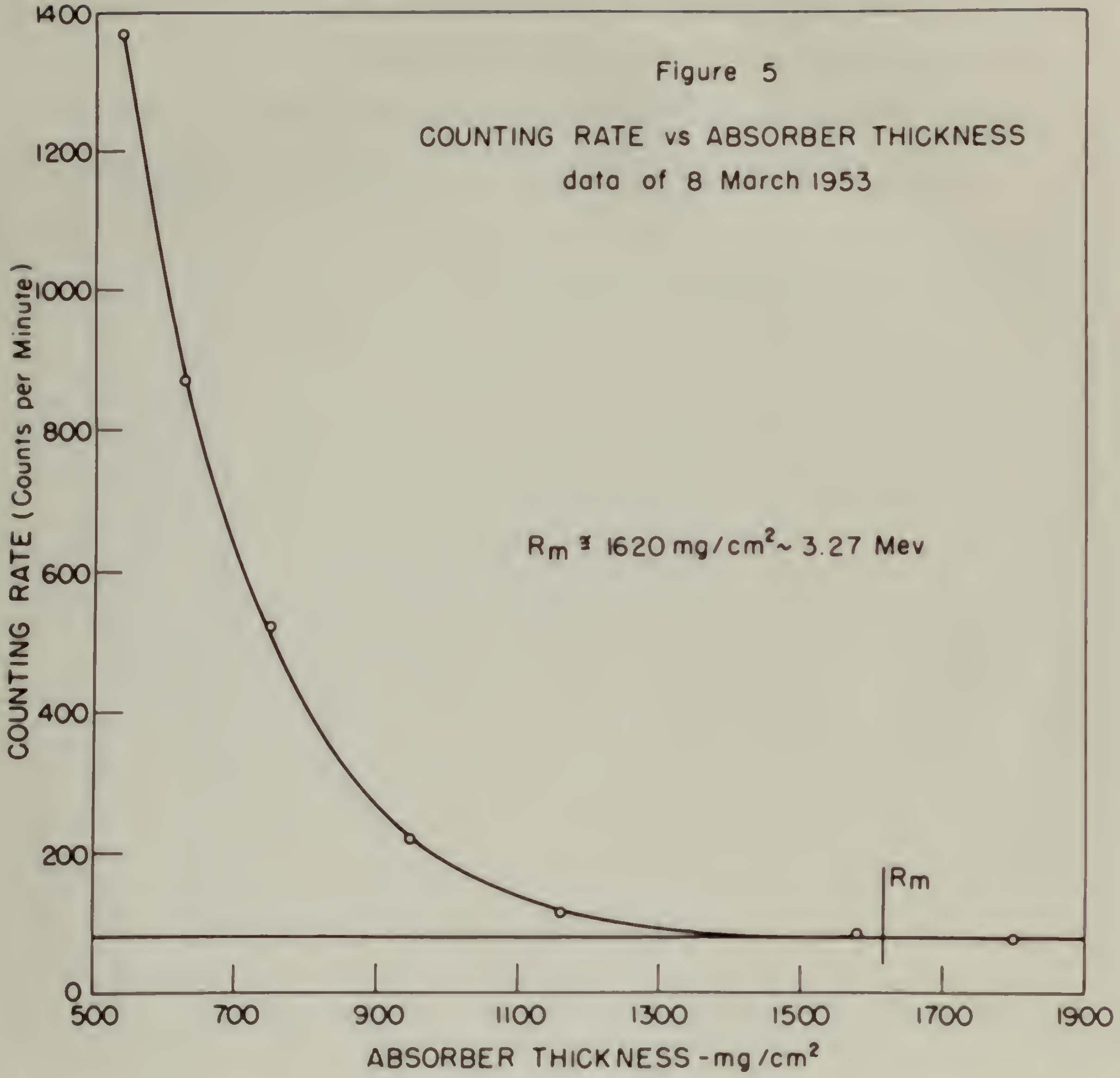


Figure 5

COUNTING RATE vs ABSORBER THICKNESS
data of 8 March 1953

$R_m \cong 1620\ \mathrm{mg/cm^2} \sim 3.27$ Mev

the 59 hour isotopes (Figs. 6, 7). The value obtained by this method for the 17.5 day isotope agrees with that found by the maximum range measurement stated above. The curve obtained for the 26 hour isotope (Fig. 8) was concave toward the origin and could not be treated by this method. A detailed discussion of the method and theory involved is contained in Chapter I of reference 22.

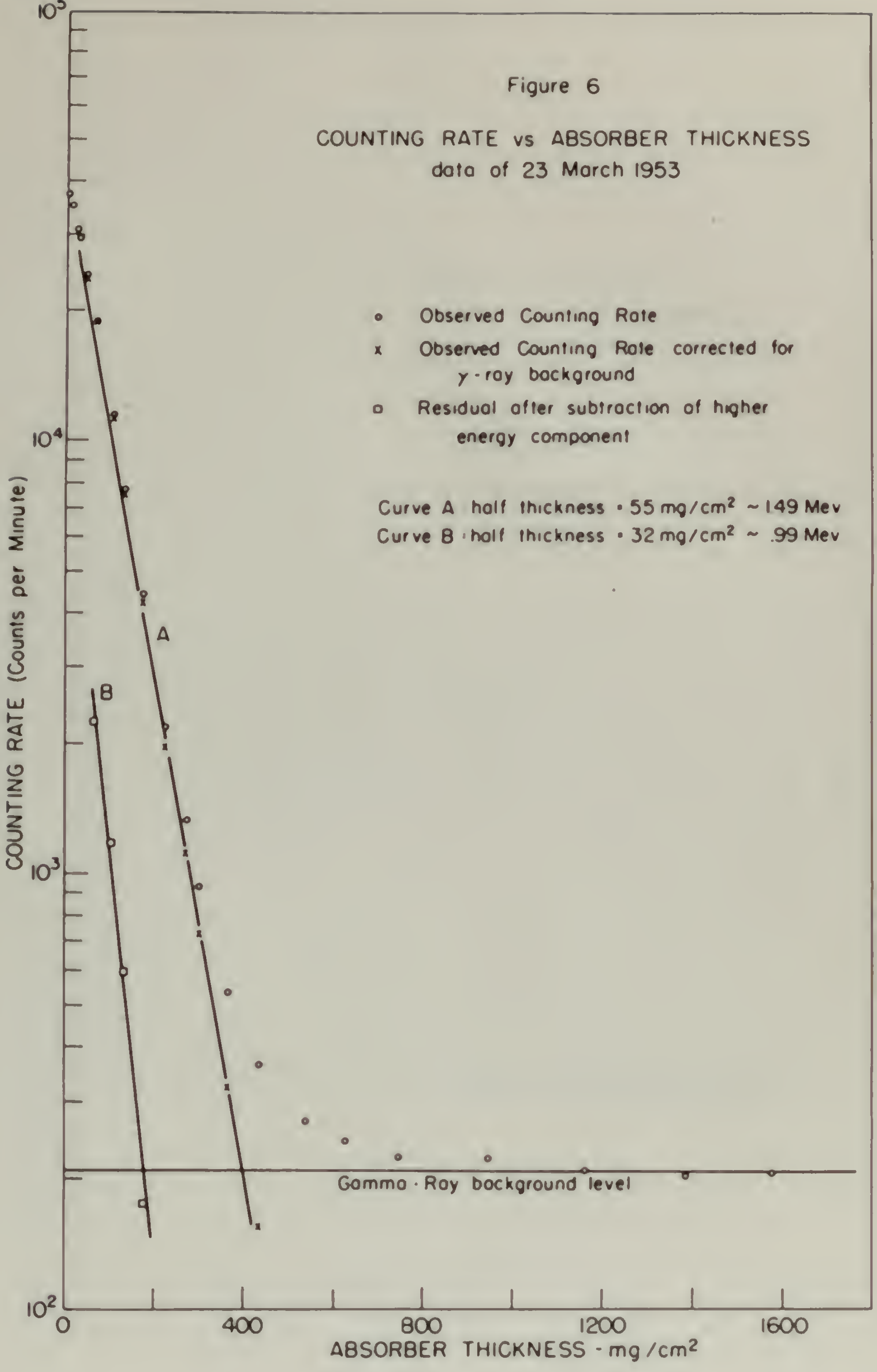


Figure 6

COUNTING RATE vs ABSORBER THICKNESS
data of 23 March 1953

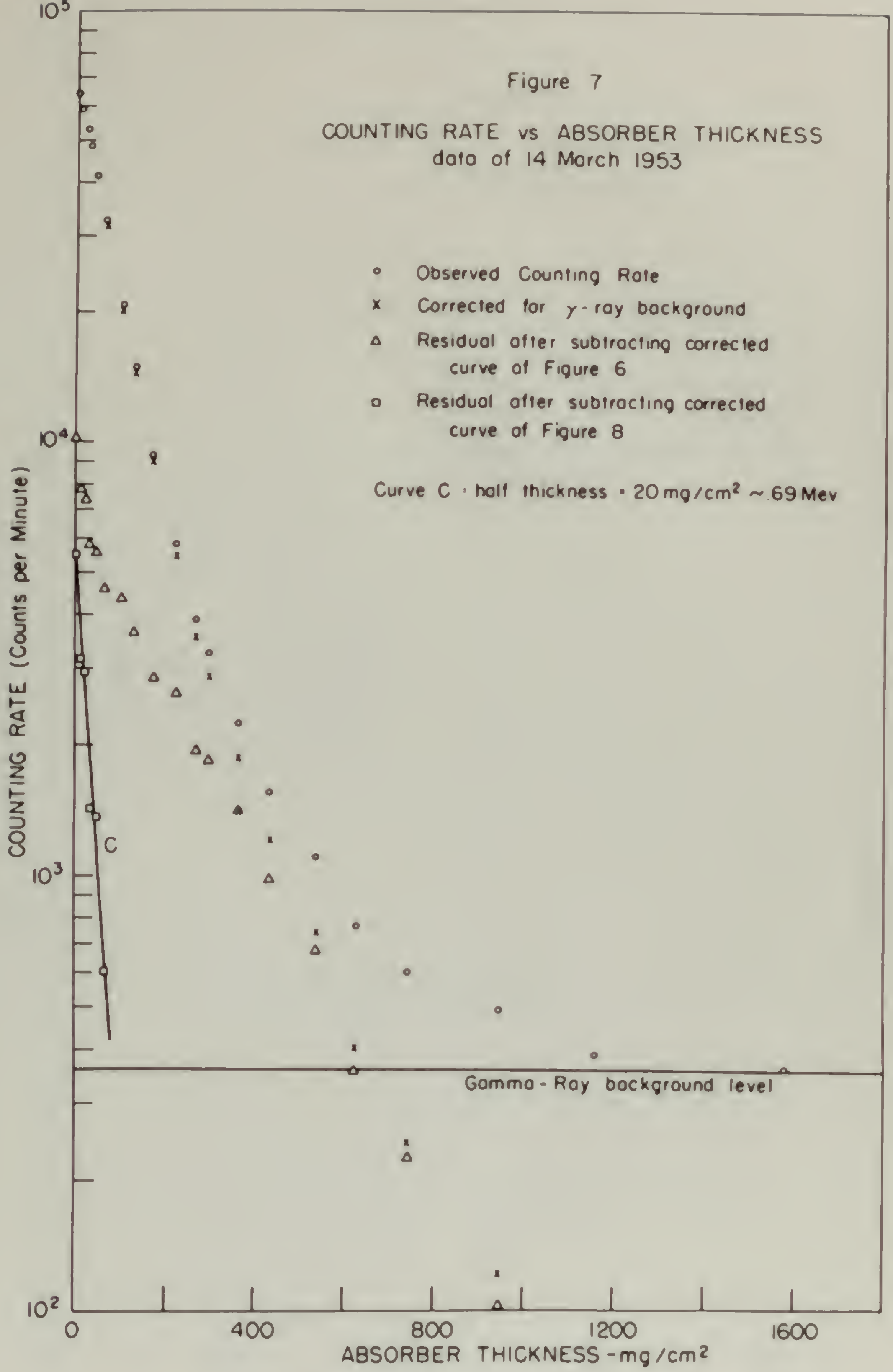


Figure 7

COUNTING RATE vs ABSORBER THICKNESS
data of 14 March 1953

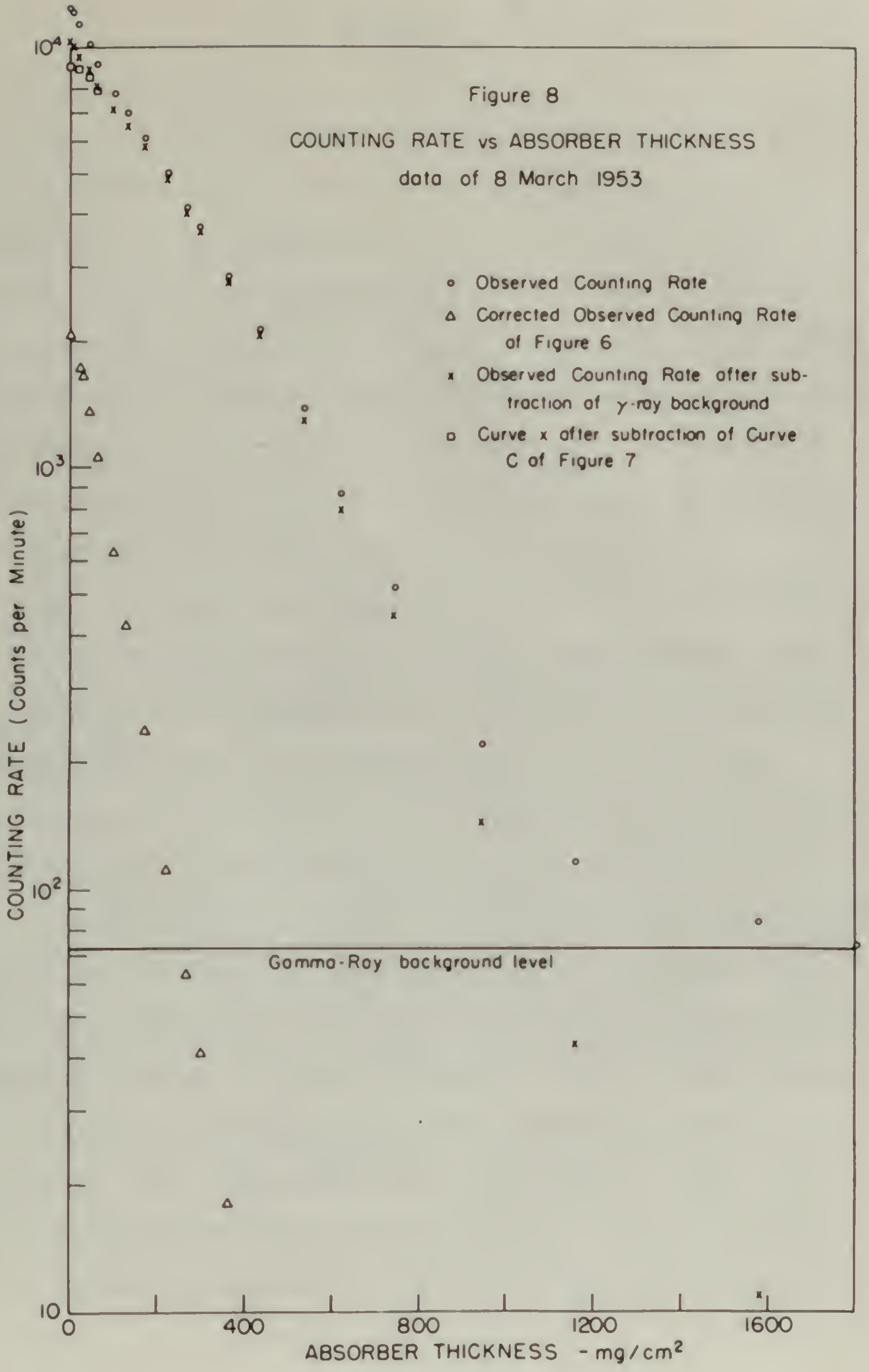


Figure 8

COUNTING RATE vs ABSORBER THICKNESS

data of 8 March 1953

V. RESULTS

1. The longest-lived isotope present in the mixture was detected by only the 4π counter. The half life was determined to be 82.9 ± 9.2 days and by the filtering method described in Section IV A, the maximum β energy was found to be 0.11 Mev $> E_{max} >$ 0.02 Mev. Since this isotope was not detected with the coincidence counter it is assumed to be a pure negatron emitter. This nuclide is believed to be As^{73} due to the close agreement with the reported characteristics of that isotope.(11)

2. With 4π, coincidence, and end window counters a half life of approximately 17.5 days was resolved. Two β energies of this nuclide were determined by absorption measurements to be 0.99 Mev and 1.49 Mev. The close agreement of these results with those previously reported(9,11,19) seem to justify the assumption that this isotope is As^{74}.

3. End window β counter measurements using an absorber thickness of 224 mg/cm^2 indicated that no isotopes were present having β energies $>$ 0.7 Mev and half lives in the range 16.7 days $> T_{1/2} >$ 26 hours. Using lesser amounts of absorber a half life of approximately 59 hours was resolved. The only other half lives found to have this approximate value were 70.1 ± 2.8 hours from 4π counter data and 48.2 ± 1.2 hours from coincidence measurements.

The latter value combined with the energy limitation previously specified justifies identification of this isotope as As^{71}.(10,11,12) The longer half life values obtained from 4π and end window counter measurements indicate that there is also present a negatron emitter having a $T_{1/2}$ longer than 70 hours with energy < 0.7 Mev. The 70 hour half life determined from 4π counter measurements is believed due to a mixture of As^{71} and As^{77} assuming that the single reported value for the half life of the latter(20) is in error. This apparent discrepancy is worthy of future study.

4. From data of the 4π and end window β counters a component of half life 25.8 ± 0.2 hours with a maximum β energy of 3.25 Mev was determined. A γ-ray energy of 0.85 Mev with half life approximately 29 hours was found from measurements made with the γ-ray scintillation spectrometer. These results confirm previously reported values(9,11,19) and identify this isotope as As^{72}. This half life determined from coincidence measurements was 21.7 ± 0.2 hours.

5. There was no indication of the presence of the 52 minute As^{70} isotope(9) in the mixture. In addition, since no γ-ray energies > 0.85 Mev were resolved it was

apparent that the 27.6 hour As^{76} isotope having two reported γ energies > 1 Mev$^{(14, 18)}$ was not present.

6. Tabular summary of characteristics of the mixture of radionuclides determined by this investigation.

Isotope	Method of decay	Energy (Mev)	$T_{1/2}$	Thick target yield* (μc/μamp-hr)
As^{71}	β^+	0.66	48.2 ± 1.2 hrs.	7.6
As^{72}	β^+ γ	3.25 0.85	25.8 ± 0.2 hrs.	64.9
As^{73}	β^-	$0.11 > E_{max} > 0.02$	88.9 ± 9.2 days	1.1
As^{74}	β^+ β^-	0.99, 1.49	17.82 ± 0.13 days	5.2
As^{76}		Not present in the mixture		
As^{77}	β^-	< 0.7	> 70 hours	$5 <$ yield < 15**

* The thick target yield values specified apply if the deuteron beam current was exactly 36 μamps and if the arsenic separation efficiency was 100 percent. Yield values quoted are based on β counting only and do not include orbital electron capture.

** Based on ratios of total β to β^+ counting rates.

BIBLIOGRAPHY

1. Sagane, R.: Phys. Rev. 54, 149 (1938)
2. Sagane, R.: Phys. Rev. 55, 31 (1939)
3. Curtiss, B. R. and J. M. Cork: Phys. Rev. 53, 681 (1938)
4. Sagane, R., S. Kojima, G. Miyamoto, and M. Ikawa: Proc. Phys-Math. Soc. of Japan 21, 660 (1939)
5. Thornton, R. L.: Phys. Rev. 49, 207 (1936)
6. Walke, H.: Phys. Rev. 52, 669 (1937)
7. Sagane, R., G. Miyamoto, and M. Ikawa: Phys. Rev. 59, 904 (1941)
8. Glendenin, L. E.: Nucleonics 2, No. 1, 12 (1948)
9. Hopkins, H. H., Jr. and B. B. Cunningham: Phys. Rev. 73, 1406 (1948)
10. Hole, N.: Arkiv Mat. Astron. O. Fysik 36A, 1 (1948)
11. McCown, D. A., L. L. Woodward, and M. L. Pool: Phys. Rev. 74, 1315 (1948)
12. Bracher, D. F. and A. R. Crathorn: Nature 169, 364 (1952)
13. Johansson, S., Y. Cauchois, and K. Siegbahn: Phys. Rev. 82, 275 (1951)

14. Bair, J. K. and F. Maienschein: Phys. Rev. 81, 653 (1951)

15. Strait, E. N., D. M. Van Patter, and W. W. Buechner: M.I.T. Progress Report, January 1950

16. Jensen, E. N., R. T. Nichols, and J. Clement: Phys. Rev. 81, 143 (1951)

17. Shure, K.: M.I.T. Progress Report May 1951, and Thesis.

18. Mims, W. and H. Halban: Proc. Phys. Soc. (London) A64, 311 (1951)

19. Mei, J. Y., A. C. G. Mitchell, and C. M. Huddleston: Phys. Rev. 79, 237A, 19 (1950)

20. Steinberg, E. P. and D. W. Engelkemeir: NNES-9, Paper 54 (1950)

21. Brownell, G. L., E. V. Bachofen, E. F. White, and J. W. Irvine, Jr.: Progress Report May 1953, Contract AT(30-1)-952

22. Goodman, C.: "Introduction to Pile Theory", Chap. 1, 2nd edition, Addison-Wesley Press, Cambridge, Mass., 1952

23. Butement, F. D. S.: Nature 165, 149 (1950)

24. Brightsen, R. A., Shure, K., Fisher, C., and C. D. Coryell: Phys. Rev. 81, 298A (1951)

APPENDIX I

THE 4π COUNTER

APPENDIX I

Table of Contents

Section A. Description of the Counter 31

B. Applicability to Absolute β Counting . . . 38

C. Important Aspects of Source Preparation 41

D. Preparation of Source Mounting Film . . . 43

E. Conducting Layer for Source Mounting Film 46

F. Preparation and Precipitation of Source Material 50

G. Technique Used in Absolute Counting . . . 56

H. Important Characteristics of 4π Counter 69

Bibliography 71

APPENDIX I

List of Figures

Figure No.	Title	Page
1	Photograph of disassembled counter	33
2	Detailed drawing of counter construction . .	34
3	General drawing of assembled counter	35
4	Photograph of counter with electronic . . . equipment connected for normal operation	40
5	Graph of color and intensity of reflected light vs film thickness.	45
6	Photograph of apparatus used in metallic evaporation.	48
7	Vacuum trap arrangement for pipette cleansing	54
8	P^{32} counting rate vs discriminator voltage for various electronic gains (counter voltage constant).	58
9	Co^{60} counting rate vs discriminator voltage for various electronic gains (counter voltage constant).	59
10	P^{32} counting rate vs discriminator voltage for various counter voltages (electronic gain constant)	60

11 Co^{60} counting rate vs discriminator voltage for various counter voltages (electronic gain constant) 61

12 Calibration curve of discriminator voltage vs discriminator dial setting 62

APPENDIX I

THE 4π COUNTER

A. Description of the Counter

The 4π solid angle counter was designed as a convenient laboratory instrument to be used for the absolute standardization of β emitters. A photographic view of the disassembled counter, Fig. 1, illustrates the important features of construction and source mounting. Detailed specifications are given in Figs. 2 and 3.

The sensitive volume of the counter is geometrically similar to that of Caswell(1) and of Borkowski(2). The counter was designed and operated as a flow counter using n butane gas rather than as a fill counter due to the fact that the former is more stable with far better reproducibility(2). Since the counter must be opened each time a source is changed, use as a flow counter which eliminates the necessity for a vacuum seal greatly simplifies the operating procedure as compared with that of a fill-type counter. The O-

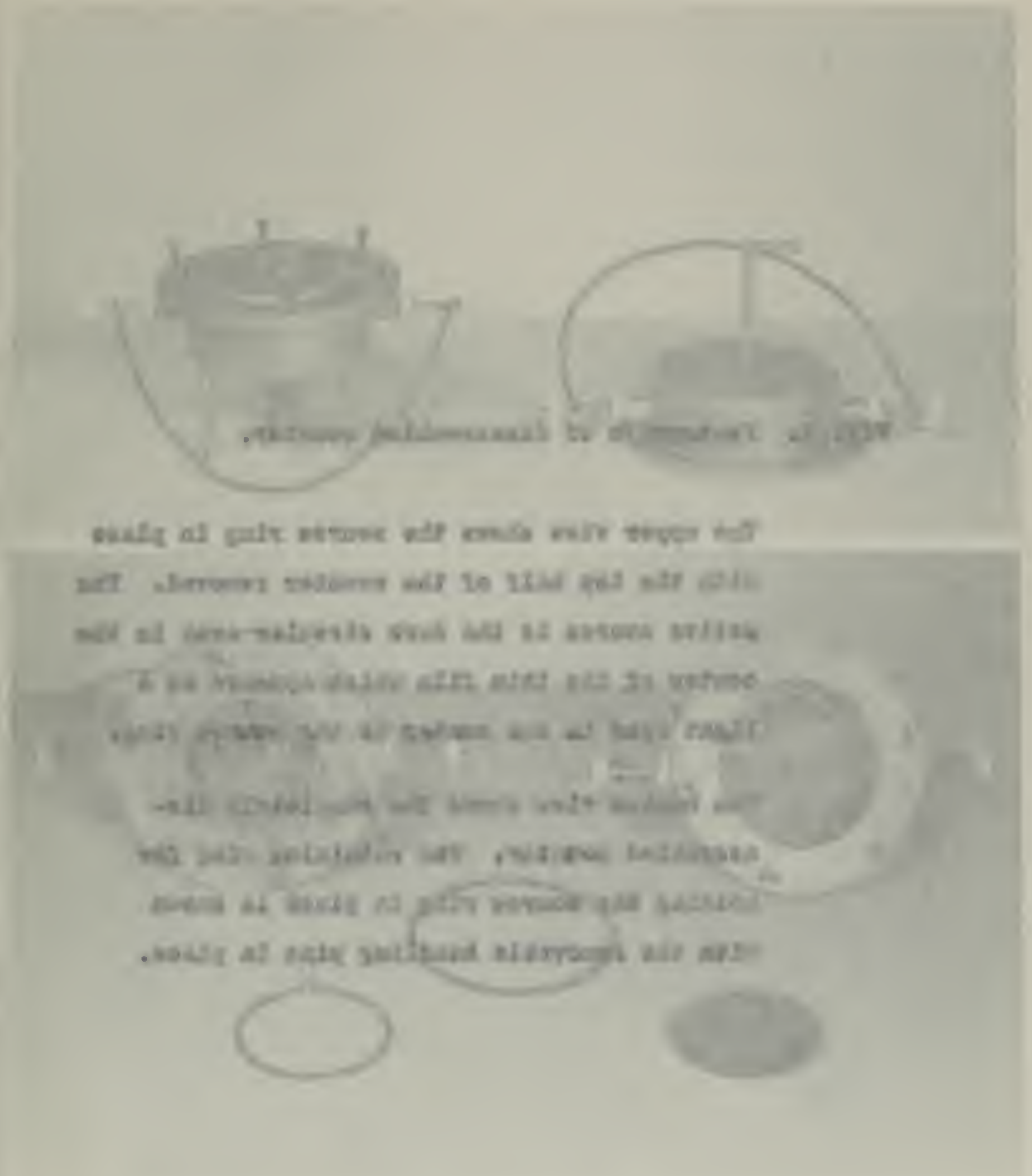
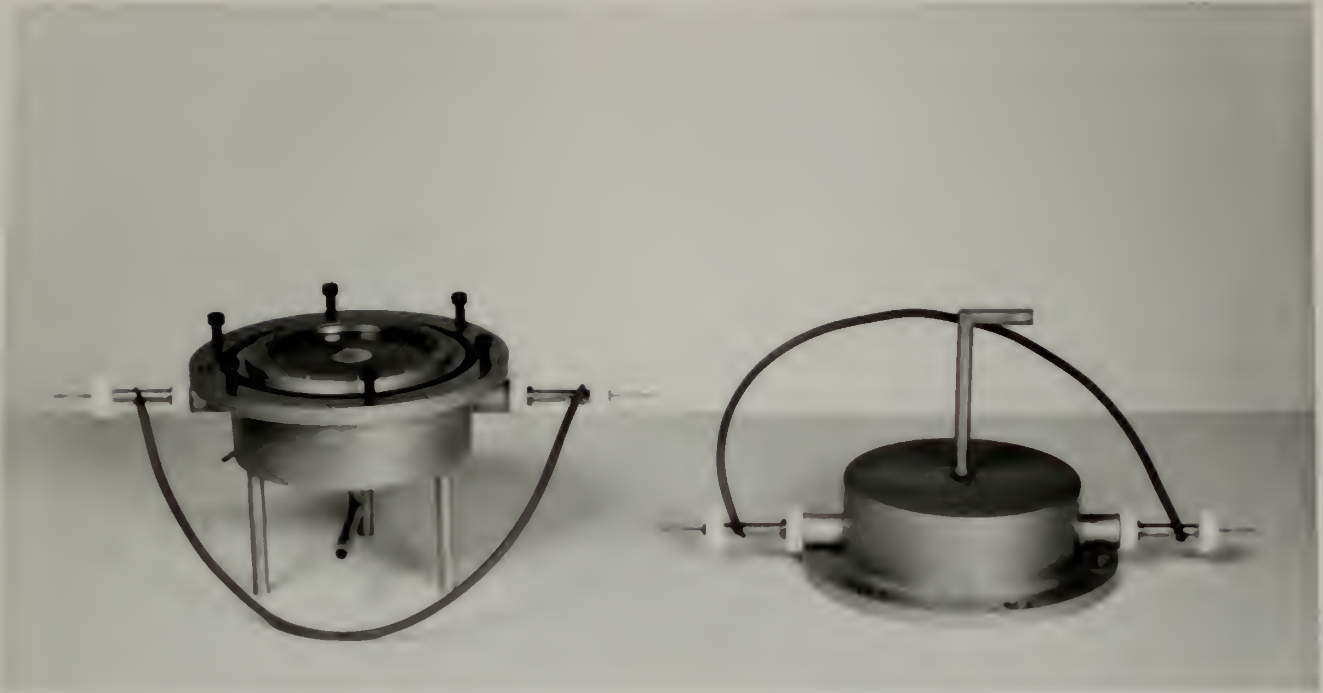
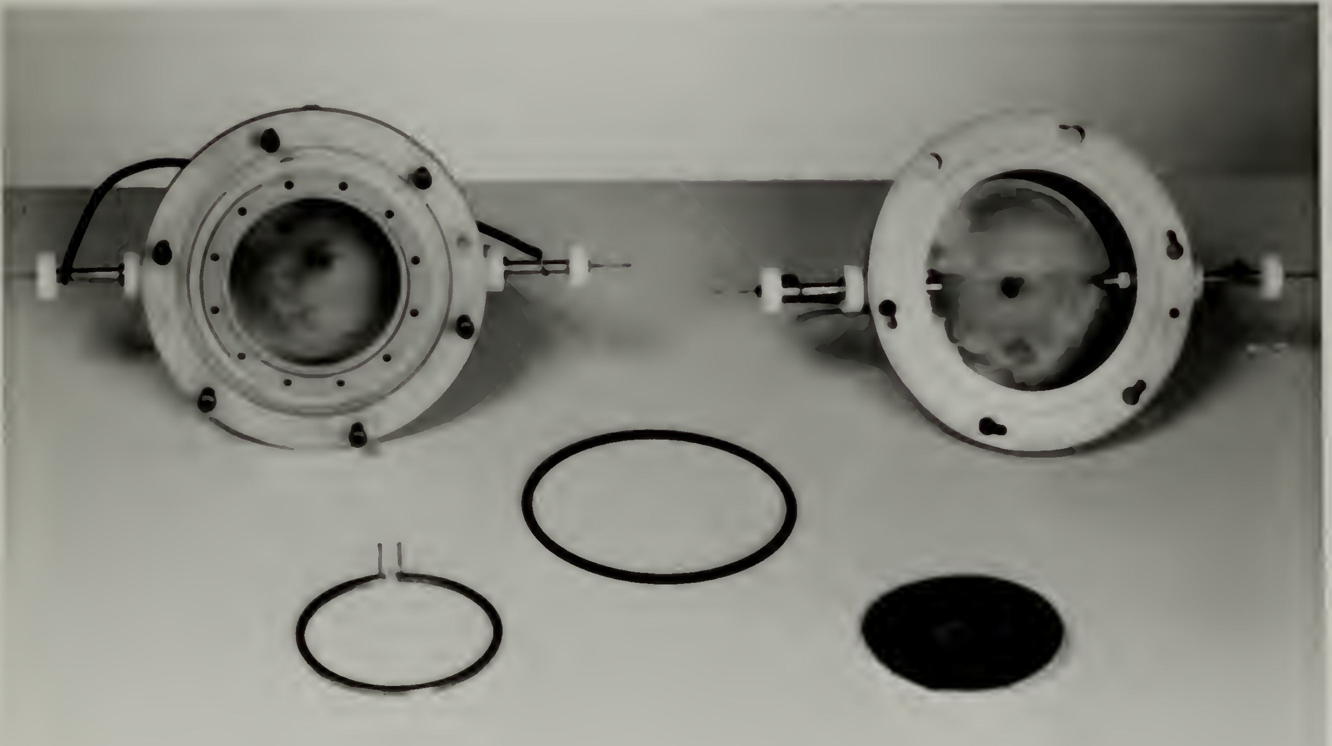


Fig. 1. Photograph of disassembled counter.

The upper view shows the source ring in place with the top half of the counter removed. The active source is the dark circular area in the center of the thin film which appears as a light area in the center of the source ring.

The bottom view shows the completely disassembled counter. The retaining ring for holding the source ring in place is shown with the removable handling pins in place.





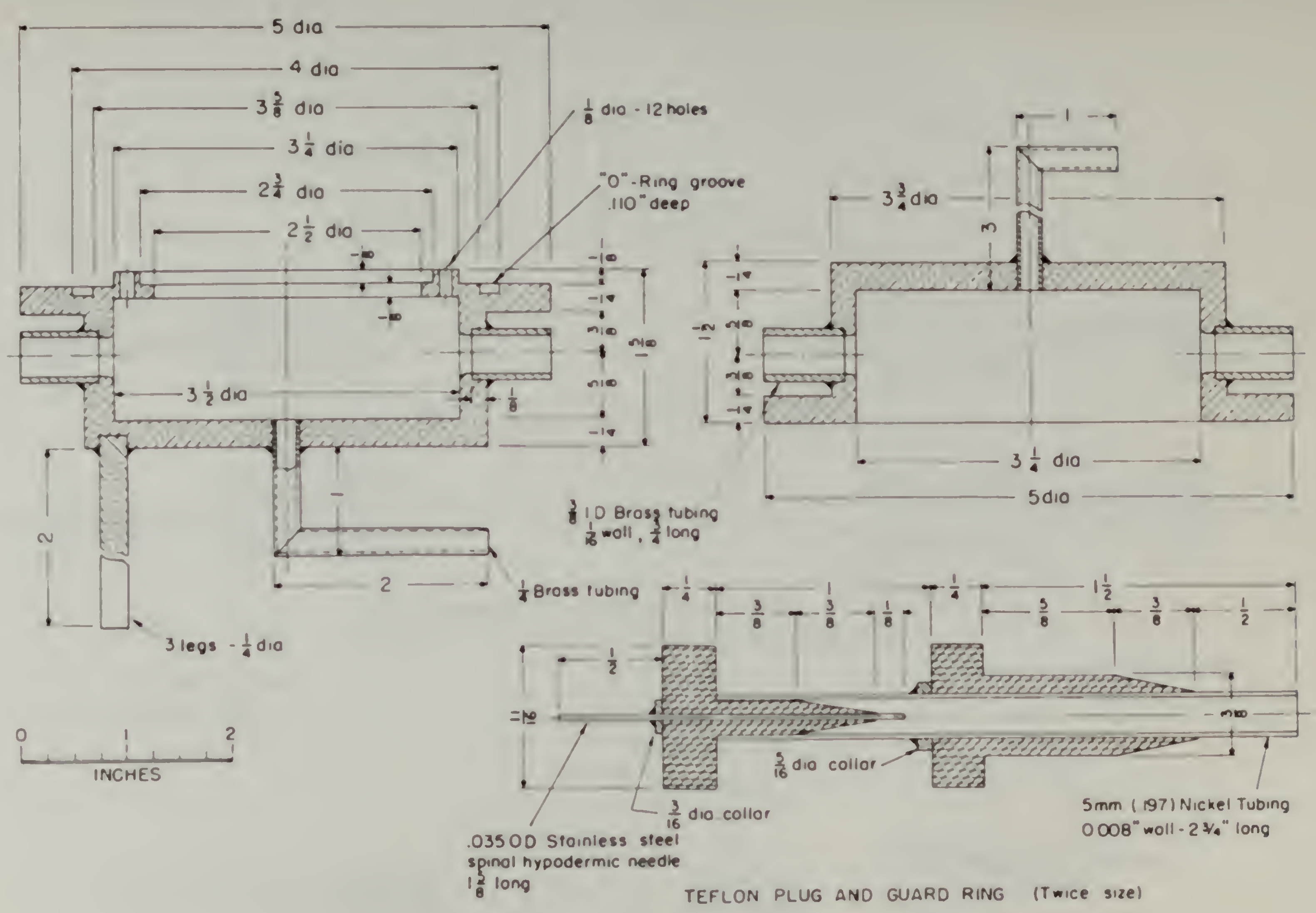


Figure 2

4 π PROPORTIONAL FLOW COUNTER

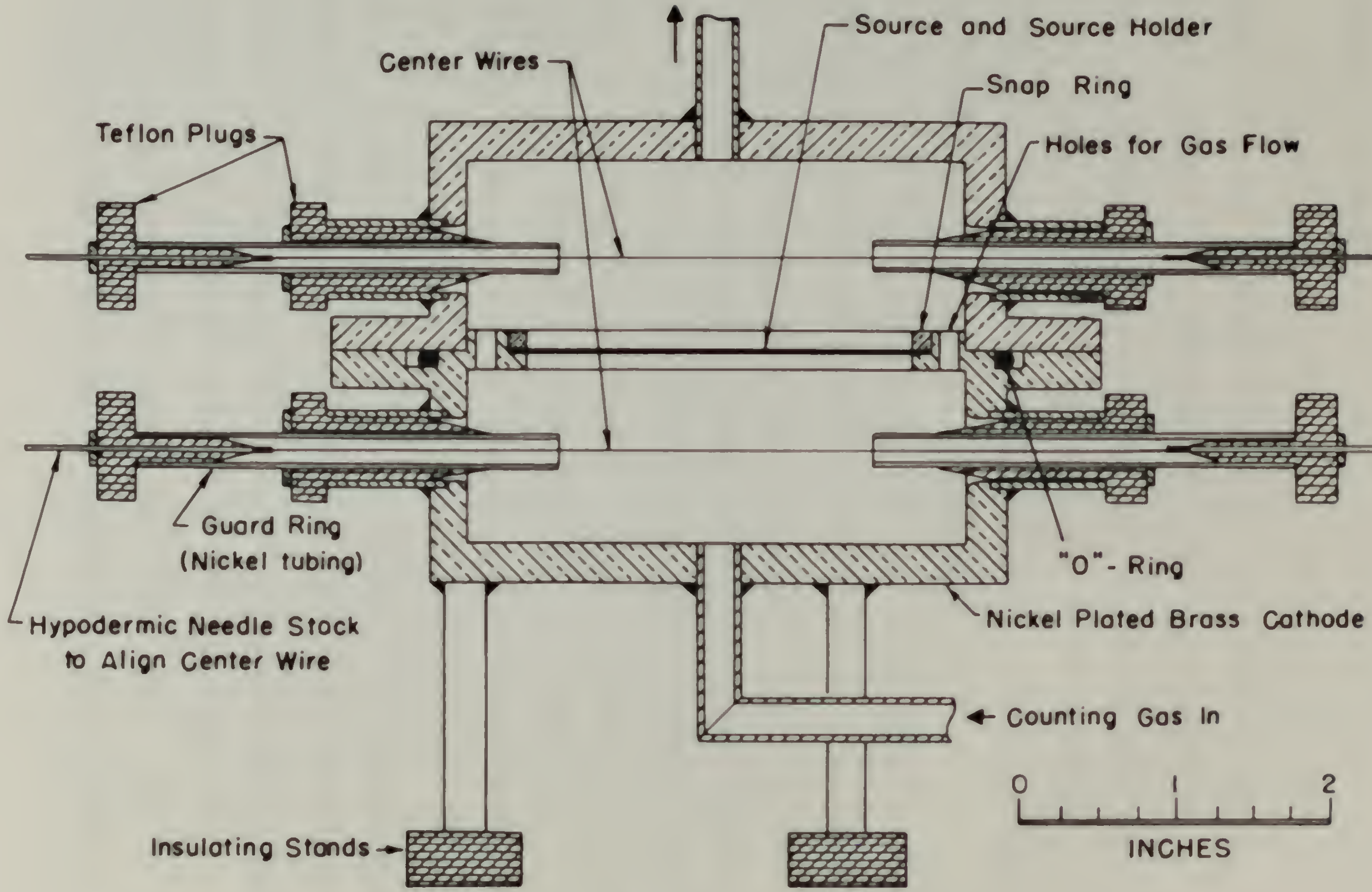


Figure 3

CROSS SECTION OF 4 π PROPORTIONAL FLOW COUNTER

ring seal is used to reduce leakage of flow gas to an absolute minimum.

Squeeze-fitted Teflon plugs are used instead of Kovar glass for counter case-to-guard ring and guard ring-to-center wire insulation, with all insulator surface leakage paths designed for approximately 5000 volts. This feature is for convenience in assembly, cleaning of the sensitive volume, and to minimize the possibility of breakage in handling.

The 1/4 inch thick counter case is machined from 5 inch brass stock, all sleeve entries to the case being silver-soldered and the entire assembly nickel-plated to facilitate cleaning. Center wires of 1 mil tungsten are aligned by 20 gauge hypodermic needle stock to which they are soldered at the extremities. Center wires are guard-ringed with the guard rings at the same high positive potential as the center wires. The tripod legs supporting the counter fit into insulating stands made of drilled polystyrene rod stock. A grounded brass shielding box contains the entire counter assembly and minimizes externally-caused electromagnetic interference. Electrical connections within the box are made with rubber-covered wire insulated for 5000 volts. All electronic connections to the shielding box are made

by coaxial cable and associated fittings. A U-tube oil-filled bubbler external to the shielding box maintains gas pressure within the counter slightly above atmospheric and avoids changes in gas concentration.

Three mil shim steel stock* provides a sturdy source mounting ring. The steel is first cut into 3 inch squares and a 5/8 inch hole is punched in the center of these squares. The 2 3/4 inch outer diameter is then obtained by use of a jig and ordinary paper shears. The prepared source ring is held in place in the counter by use of a split brass ring (similar to a piston ring).

To retain some of the pulse limiting properties of the conventional Geiger counter while eliminating many of its objectionable features, the 4π counter is operated in the region of limited proportionality.(4, 5) The counter is operated at a well regulated 4300 volts with the cathode 2500 volts below ground and center wires and guard rings 1800 volts above ground to minimize corona and "spurty" noise effects. This operating point is approximately 500 volts above the beginning of a counting rate plateau which is better than 0.6 percent per 100 volts. The center wire output is fed

---

* Obtainable from Ward Steel Co., Arlington, Mass.

to a Model 100 amplifier through a cathode follower preamplifier (Atomic Instrument Co. Model 204-B) with the amplifier output driving an M.I.T. Model 400-R decade scaler. The counter with associated electronic equipment connected for normal operation is illustrated in Fig. 4.

B. Applicability to Absolute β Counting

If a counter is built which collects particles emitted from a source in all directions, it has many advantages for measurement of absolute activity. First, since all particles emitted from the source are counted, a direct measurement of the β disintegration rate is made without the need for precise knowledge of the solid angle with its accompanying scattering problems. Also, since the "efficiency for β particles" is now 100 percent, much smaller and thinner sources may be prepared thus reducing the self-absorption considerations.(8)

In the 4π counter, any β particle which produces an ion pair outside the source and source mounting will be counted unless this ion pair is formed in a region of low enough intensity that recombination occurs prior to

Fig. 4. Photograph of counter with electronic equipment connected for normal operation.

The insulating polystyrene mounts are visible within the brass shielding box and the oil-filled bubbler is shown on the outside lower left corner.

initiation of the Townsend avalanche. Consideration of the geometry of and the fields existing in the sensitive volume indicates a very small probability for counting losses due to this effect.(1)

Any ionization produced by internal conversion electrons, branched spectra, γ-ray spectra, and electrons produced in the counter walls or in the gas will merely add to the total ionization per disintegration and will therefore be counted as a single pulse. This is also true of annihilation radiations and this fact makes the 4π solid angle method valid for the assay of positron emitters.

Deviations from 100 percent absolute efficiency will be due only to (a) absorption in the source and source mounting film, (b) areas of low field intensity mentioned above, and (c) resolving time losses.

C. Important Aspects of Source Preparation

The preparation of a thin source is the most difficult problem involved in the practical use of the 4π counter. It is essential that the source be quite thin and uniform for any isotope emitting soft β particles. The

chemistry involved in preparing uniform thin sources varies with the element involved. When a sample is simply allowed to dry, the active material has a tendency to crystallize out as one or more large particles or to dry in a thick ring of small crystals around the edge of the drop. Use of an infrared lamp speeds evaporation and reduces the tendency to "cluster" in every case attempted. It has been empirically determined that counting losses due to self-absorption can be neglected if the maximum solid content of the source is $\leq$ 5 μgm for β energies $\geq$ 0.6 Mev, but for β energies $\leq$ 0.4 Mev solid content of the source should not exceed 0.1 μgm. These approximate values are based on a total pipetted source volume of 0.085 ml. Within the specified limits, self-absorption losses are negligible compared to losses in the conductive layer on the source mounting film. Self-absorption can never be entirely eliminated by continued reduction of total solids since there is a finite particle size which the material must assume upon precipitation. It has been shown that below a certain very small concentration, a decrease in solids does not increase the observed counting rate. Also, a slight increase in solids above this value does not decrease the observed counting rate of a source.(3)

D. Preparation of Source Mounting Film

A solution made by dissolving 5 grams of stick parlodion* in 85 ml of amyl acetate was found to produce the most durable very thin uniform films. A period of about two weeks, with frequent agitation, is required for the formation of the solution.

Thin films are made by dropping an appropriate amount of the above solution on a clean surface of distilled water. The water used should first be boiled to eliminate dissolved gases and an indicator such as phenolphthalein should be added in order to check pH. Water which is even slightly acidic seems to decrease the physical strength and life of the film produced. A room should be chosen which is as dust- and draft-free as possible and a strong light is essential for inspecting the films and the water surface.

The simplest and most expeditious method is as follows:

1. Fill an 8-10 inch diameter culture dish to overflowing with the water prepared as indicated above.

2. Express two drops of parlodion solution on the clean water surface and observe the color display under a strong white light as the film spreads.

---

* Obtainable from Central Scientific Co., N. Y.

3. When maximum color display is evident near the edges of the film, drop the prepared source ring horizontally from a height of about 1/2 inch onto the center of the floating parlodion film.

4. Holding one edge of the floating source ring and attached film, trim away the excess film with a very sharp knife. The ring is then slid from the surface of the water at a small angle to avoid surface tension film breakage and may be placed vertically in a drying rack.

5. Film thickness may be determined by observation of reflected color under white light and comparison with available curves which read directly in $\mu gm/cm^2$ (Fig. 5). For more accurate determination, the α thickness gauge may be used. This consists of a collimated source of polonium fastened to a movable micrometer jaw which is mounted vertically above a thin window Geiger counter. A zero reading of the end of the α particle range is made, after which the film is placed over the counter window and the measurements repeated. The distance between the two curves so obtained gives the absorption of the film in air-mm, which can be translated directly into $\mu g/cm^2$. The gauge is capable of measuring thicknesses as small as 1 $\mu g/cm^2$ with less than 10 percent error.

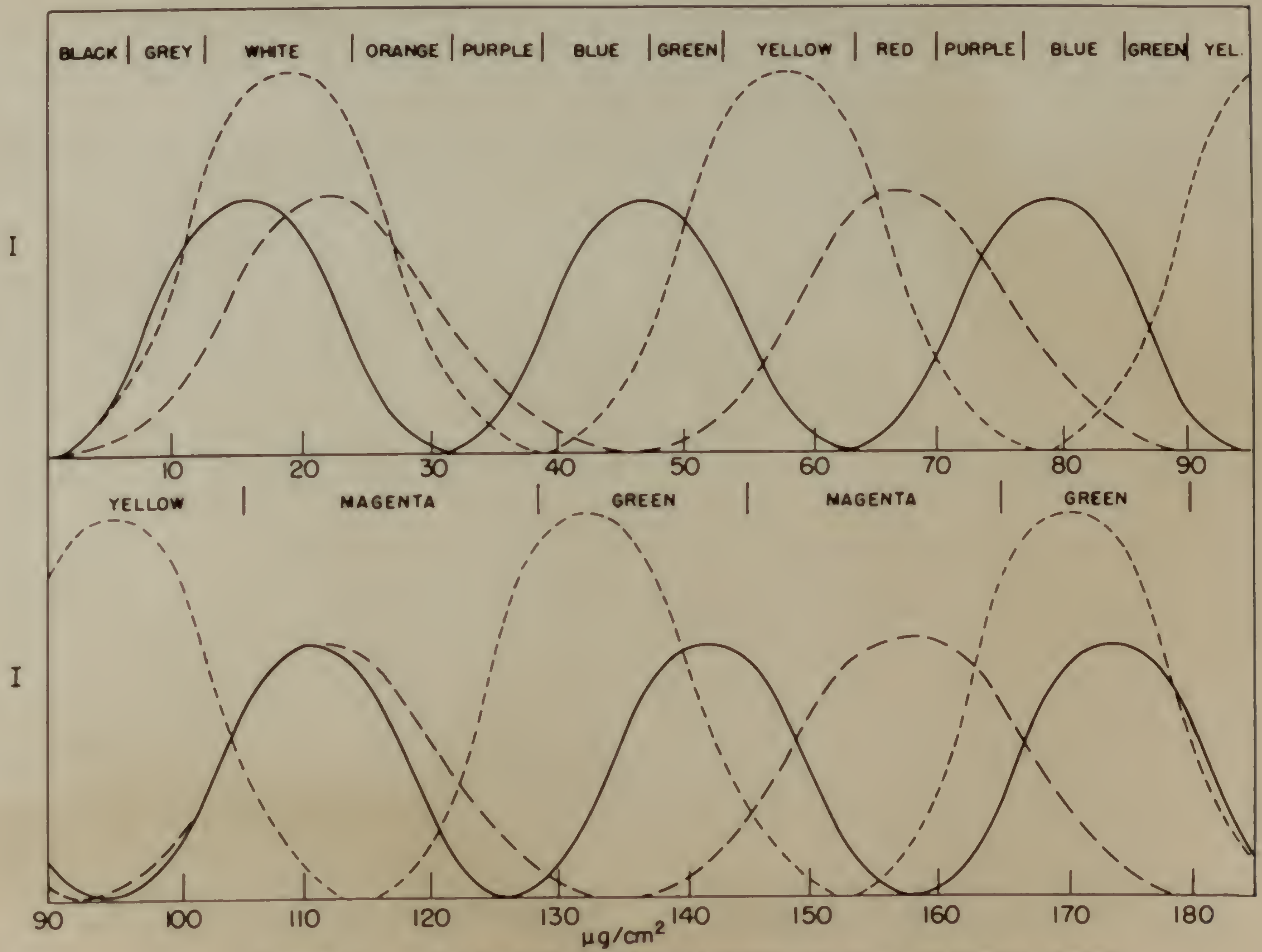


Figure 5 INTENSITY OF REFLECTED LIGHT vs FILM THICKNESS

E. Conducting Layer for Source Mounting Film

For absolute measurements, it is essential that the collecting field within the counter not be distorted by the dielectric-covered hole in the source ring. A thin conducting metallic layer which covers the entire source support may be evaporated from a heated tungsten filament, the evaporation being performed in a vacuum of approximately 1 micron. To insure electrical contact with the counter case, the layer should be deposited on the side of the source ring opposite that to which the parlodion film adheres. The apparatus used for metallic evaporation is illustrated in Fig. 6 where the method of supporting the source ring described below is clearly visible. Since the greatest danger of film breakage occurs in the metallic evaporation process, this operation should be performed prior to pipetting the active source material.

For source solutions which do not contain hydrochloric acid, aluminum produces a suitable conducting layer and the following procedure is recommended.

1. Place the prepared source ring horizontally atop a length of 50 mm diameter glass or pyrex tubing which encloses the prepared tungsten filament. The

Fig. 6. Photograph of apparatus used in metallic evaporation.

The source ring lying on top of the cylindrical glass tubing under the bell jar is in proper position for aluminum evaporation. Tubing which carries cooling water for the filament electrodes is visible to the left of the bell jar.

Fig 6

source ring should be approximately 7 cm above the filament for a vacuum of 1 μ.

2. When a vacuum of 1 μ is reached, slowly increase filament current until the aluminum begins to evaporate from the filament. Observe the climb of deposited aluminum on the glass tube and when it reaches the top of the tube, shut off filament current. This procedure results in uniform conducting layers of from approximately 10 to 15 $\mu g/cm^2$ in thickness.

The hydrochloric acid in many source solutions will interact with the aluminum surface of the source mounting and frequently causes a decrease in counting efficiency. In such cases a thin layer (15-20 $\mu g/cm^2$) of gold produces a suitable conducting surface. A standard microscope slide placed at the same vertical distance above the evaporating filament as the source mounting film and coated simultaneously with the source film provides a measurement of the thickness of gold. A resistance across the length of the slide measuring between 50 and 200 megohms indicates a thickness of gold between 15 and 20 $\mu g/cm^2$.(3)

For the measurement of isotopes having β energies greater than 1 Mev, aluminum foil of 0.1 mil thickness*

---

* Obtainable from Frank H. Caffin and Son, 22 Elm St., Hyde Park, Mass.

may be used instead of the evaporated metallic layer with no detectable counting loss. A fine mist of distilled water is deposited on the prepared source ring by use of an ordinary bulb type atomizer. The 0.1 mil foil is then laid over the moistened source ring, carefully brushed flat with a fine camel's hair brush, and the excess trimmed off with scissors. If the above is carefully performed the foil is then inseparable from the source ring and parlodion film.

No conducting layer need be applied to the source mounting if a high degree of accuracy is not required. Elimination of the metallic layer results in counting losses of from approximately 1 percent to 3 percent depending on the maximum β energy of the isotope used. For example, the observed counting rate from several non-conducting P^{32} sources increased by 2 ± 0.5 percent with the addition of either foil or evaporated aluminum conducting coatings.

F. Preparation and Precipitation of Source Material

1. Isotopes emitting β particles of $\geqslant$ 0.6 Mev.

The best method found so far consists of adding

a small amount of Bentonite, a colloidal mud, to the pipetted drops of source solution. The source ions are adsorbed on the Bentonite which dries in a fairly uniform layer of fine particles. Microscopic observation of sources prepared in this manner yields a typical size of 1 micron for the largest particles, i.e., 0.1 mg/cm^2 for material of density 1.$^{(1)}$ The layer is much more uniform if instead of simply allowing the source to dry, an infrared lamp is used to decrease evaporation time.

Using the highest specific activity source material available to minimize source solid content, a solution of from 1 to 1.5 μc/ml is prepared. This yields approximate counting rates from 55×10^3 to 82.5×10^3 dpm per 25λ of active material. Since the resolving time of the counter is approximately 20 μsec, this range of activities limits resolving time losses to ≤ 2 percent.

In solutions of materials of high specific activity, considerable losses may be caused by adsorption of the active constituents on the walls of containing vessels and pipettes used in measurement.$^{(2)}$ This effect results in a decrease of activity in solution, especially

for carrier-free materials. This loss of activity may be reduced by the addition of inactive isotopes of the same chemical form as carriers prior to preparation of the source solution. For example, a small quantity of KH_2PO_4 is used with solutions of carrier-free P^{32} and KI is used with carrier-free I^{131}. The mass of carrier which may be added is determined by the permissible solid content of the solution but a desirable ratio to make adsorption negligible is approximately 10^6 inactive atoms per active atom.

The pH of the active solution is maintained so as to keep the active atoms in solution. For some isotopes the solution should be acidic while others require a basic solution. A general rule which has few exceptions is to prepare an acidic solution if the active atom is in the cation and a basic solution if it is in the anion.(8) In all solutions, any substance added to adjust the pH must be soluble when combined with the active material in order to prevent precipitation.

The following procedure is recommended for the actual source preparation:

a. Express 25λ (0.025 ml) of active solution on the center of the 5/8 inch diameter metallic coated parlodion film. The micropipette should be rinsed twice

onto the source ring in order to remove all active material from the pipette. It has been experimentally determined that the following percentages of active material are contained in rinses of the pipette:

1st rinse — 3 percent of initial contents
2nd rinse — 1 percent of initial contents
3rd rinse — 0.5 percent of initial contents

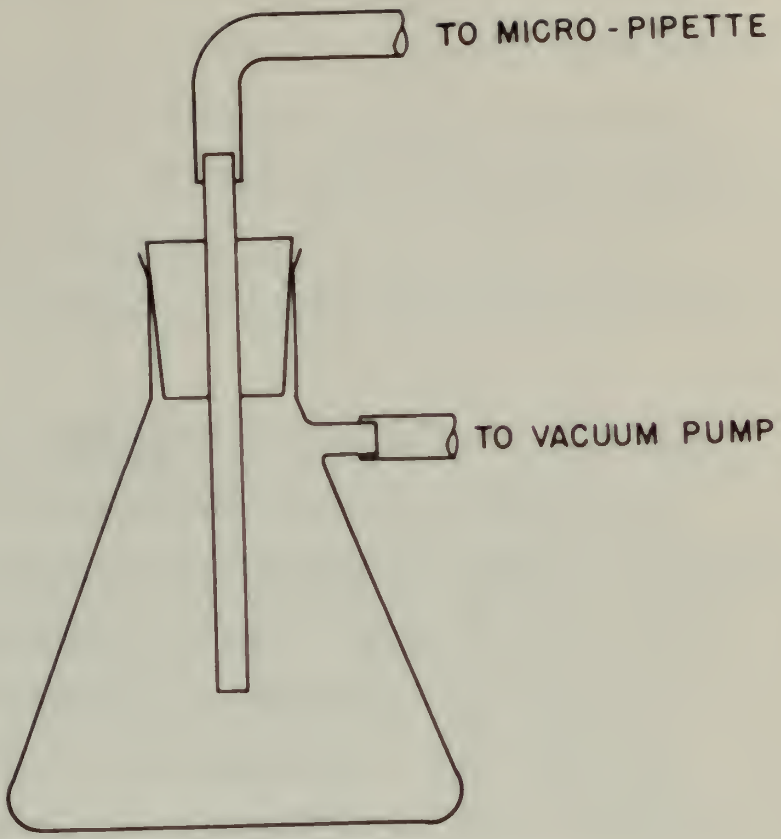
Micropipettes* used in source preparation must be calibrated with mercury since deviations of 1 percent from labeled volume are not uncommon.

b. If several sources are to be prepared at a time, the vacuum trap arrangement illustrated in Fig. 7 is indispensable. After each source (consisting of one pipette volume plus two rinses) is expressed, the pipette must be thoroughly cleaned and dried prior to preparing the next source. Using the vacuum trap arrangement the pipette can be cleaned with an inactive carrier solution followed by flushing with pure distilled water and then dried by the air stream pulled through the pipette. The total operation of cleaning and drying requires less than 3 minutes with this apparatus.

c. Express 10λ of Bentonite solution (approximately 25 mg Bentonite/ml H_2O) into the drop of source

---

* Obtainable from Radiation Counter Lab., 1844 W. 21st St., Chicago, Ill.



VACUUM TRAP ARRANGEMENT FOR CLEANING AND DRYING PIPETTES

Figure 7

solution previously formed.

d. Thoroughly disperse the Bentonite in the source solution using an air jet produced from an eyedropper which has been flame-drawn to a fine capillary point. A strong light facilitates visual observation of the mixing which is complete when the entire drop takes on a cloudy appearance.

e. The prepared source is then dried under a heat lamp.

2. Isotopes emitting β particles of $\leq$ 0.4 Mev.

In order to minimize self-absorption losses in the measurement of soft β particles extreme care must be taken in preparation of the thin source, especially if the isotope solution is a chloride which tends to form large crystals on precipitation. The following procedure, applicable to preparation of Co^{60} sources, is cited as an example.(6)

a. Using Co^{60} of high activity (approximately 1 curie/gram), dilute to proper operating range using redistilled HCl. The solid content of ordinary HCl often exceeds the solid content of the source material. The carrier concentration should be of the order of 3 mg of $CoCl_2$/liter giving a total solid content in a 25λ aliquot of 0.075 μg.

b. After pipetting the required amount of active solution on the source film, evaporate the Co^{60} to dryness as $CoCl_2$ in order to get rid of the HCl. Then add a drop of water to the evaporated material to redissolve the $CoCl_2$.

c. NH_3, introduced as NH_4OH in a beaker, should be used to precipitate the cobalt which should cover the entire area of the original water drop quite uniformly.

Steps (b) and (c) above should be done in a desiccator with sodium hydroxide used as a desiccant. A Co^{60} source carefully prepared as outlined above will reduce self-absorption to the minimum value known to be obtainable at this time.

G. Technique Used in Absolute Counting

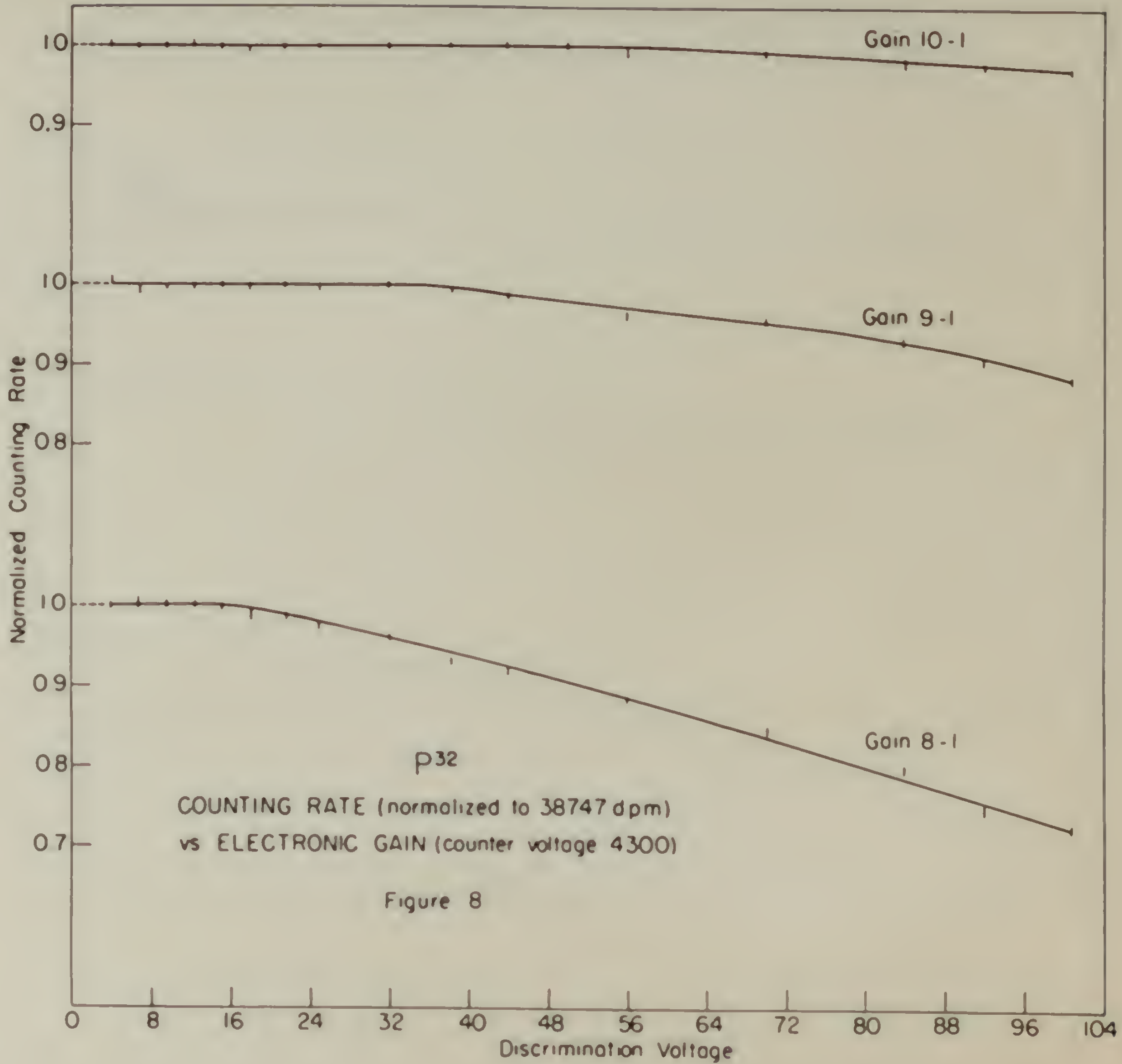
1. Sources.

Normally three sources are prepared as outlined in Section F from each solution to be counted. Comparison of counting rates of the three sources gives a measure of the precision in source preparation. With a little

practice the difference between sources due to all errors involved in preparation may be maintained at $<$ 1 percent.

2. Counting procedure.

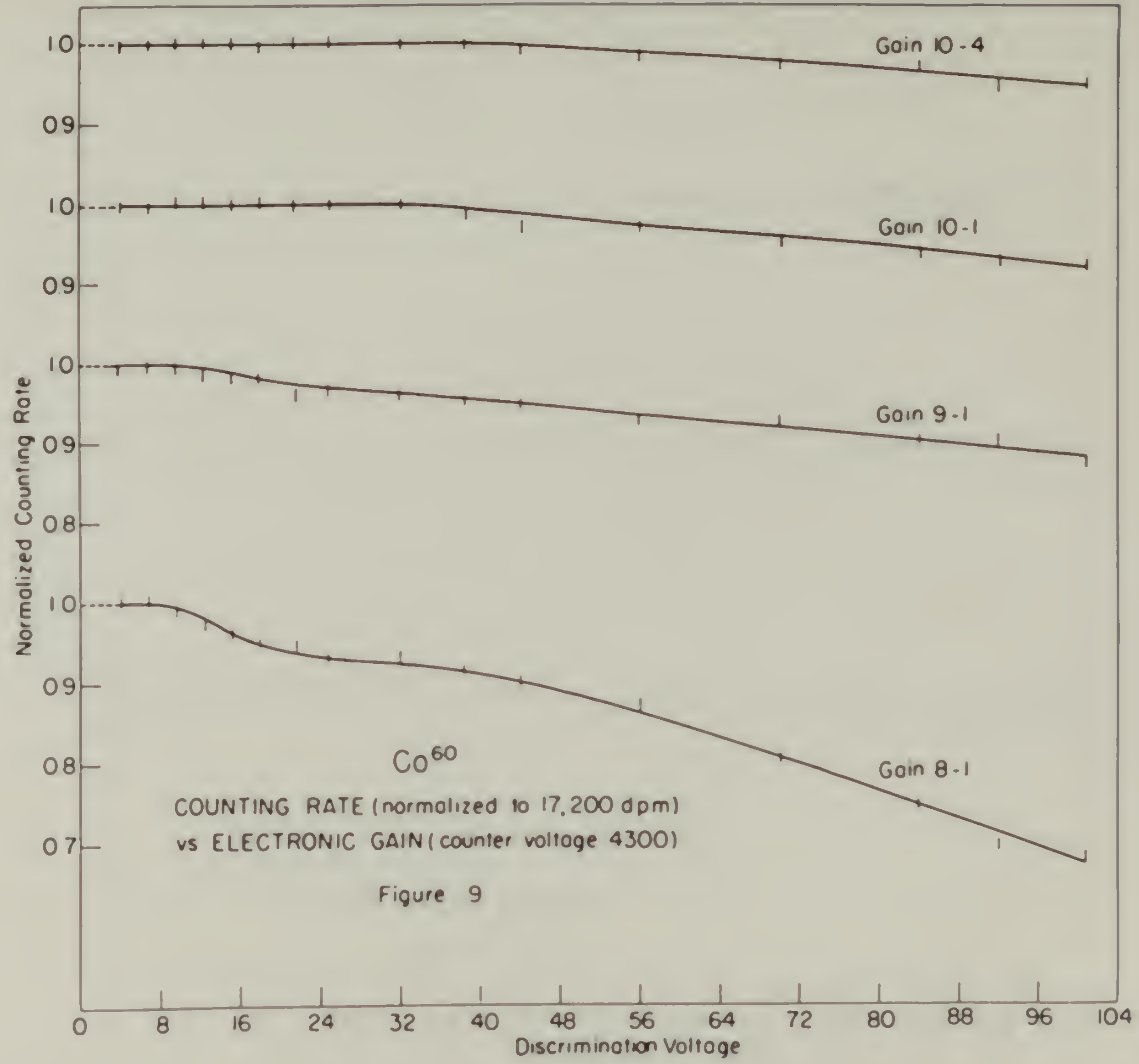
Background counts are taken before (and after if necessary) each run by inserting a plain shim steel disc in place of the source. Two comparison tests are made on all measurements made with the 4π counter. First, with counter voltage fixed at 4300 volts, integral discriminator curves are plotted for gain settings of 10:1, 9:1, and 8:1. The latter two settings each decrease electronic gain by a factor of approximately 2 (Figs. 8 and 9). Secondly, with electronic gain held constant at 10:1, integral discriminator curves are plotted for counter voltages of 3900, 4100, and 4300 volts (Figs. 10 and 11). If in both cases the discriminator curves are flat over a discriminator range of $\geq$ 10 volts (Fig. 12), we can assume that all β particles emitted into the sensitive volume are being counted. Figures 9 and 11 which are typical of Co^{60} clearly indicate the high percentage of collection and may be compared with Figs. 8 and 10 which are typical of P^{32}. The method of extrapolation to determine the true



P^{32}

COUNTING RATE (normalized to 38747 dpm)
vs ELECTRONIC GAIN (counter voltage 4300)

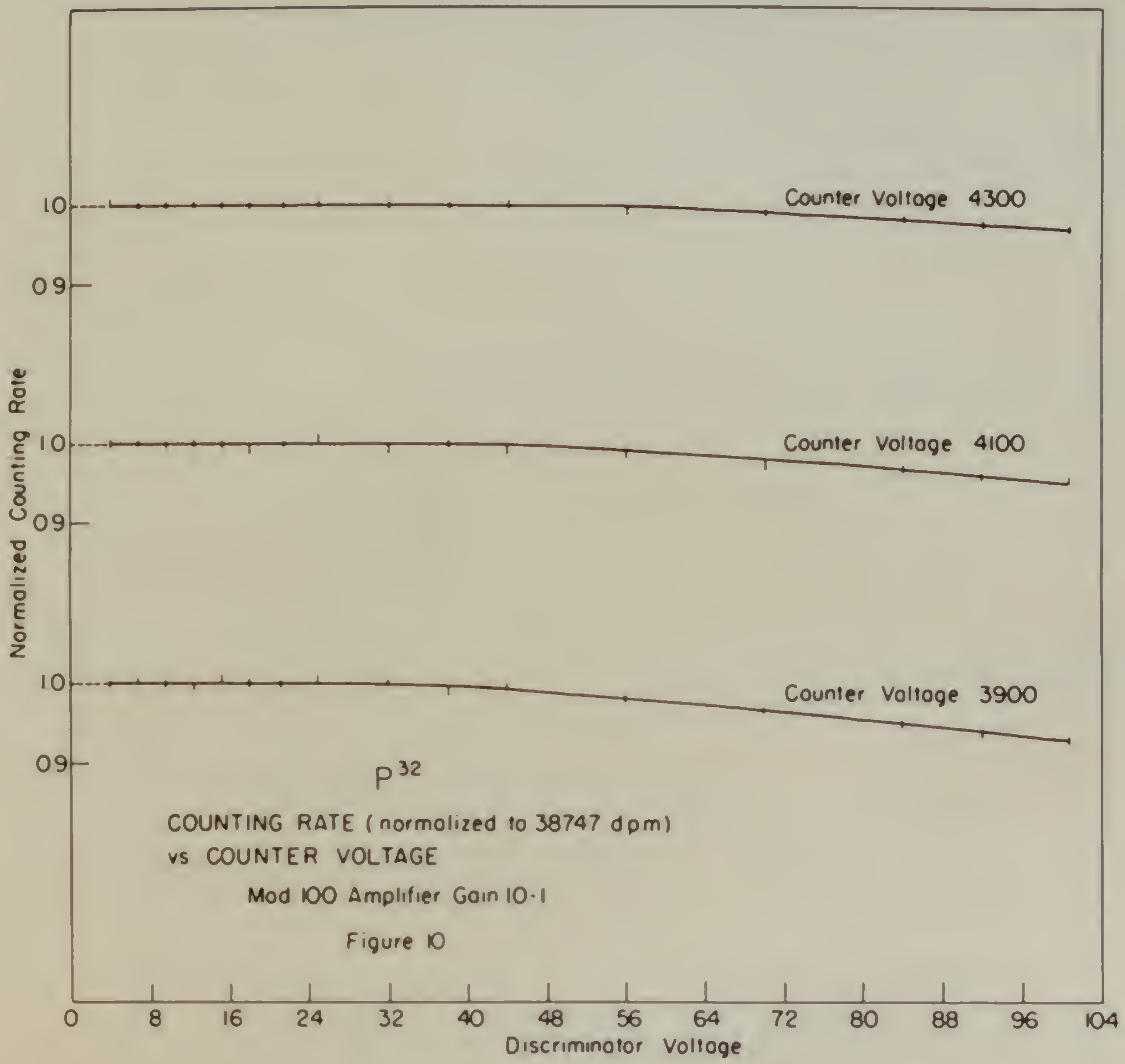
Figure 8



Co^{60}

COUNTING RATE (normalized to 17,200 dpm)
vs ELECTRONIC GAIN (counter voltage 4300)

Figure 9

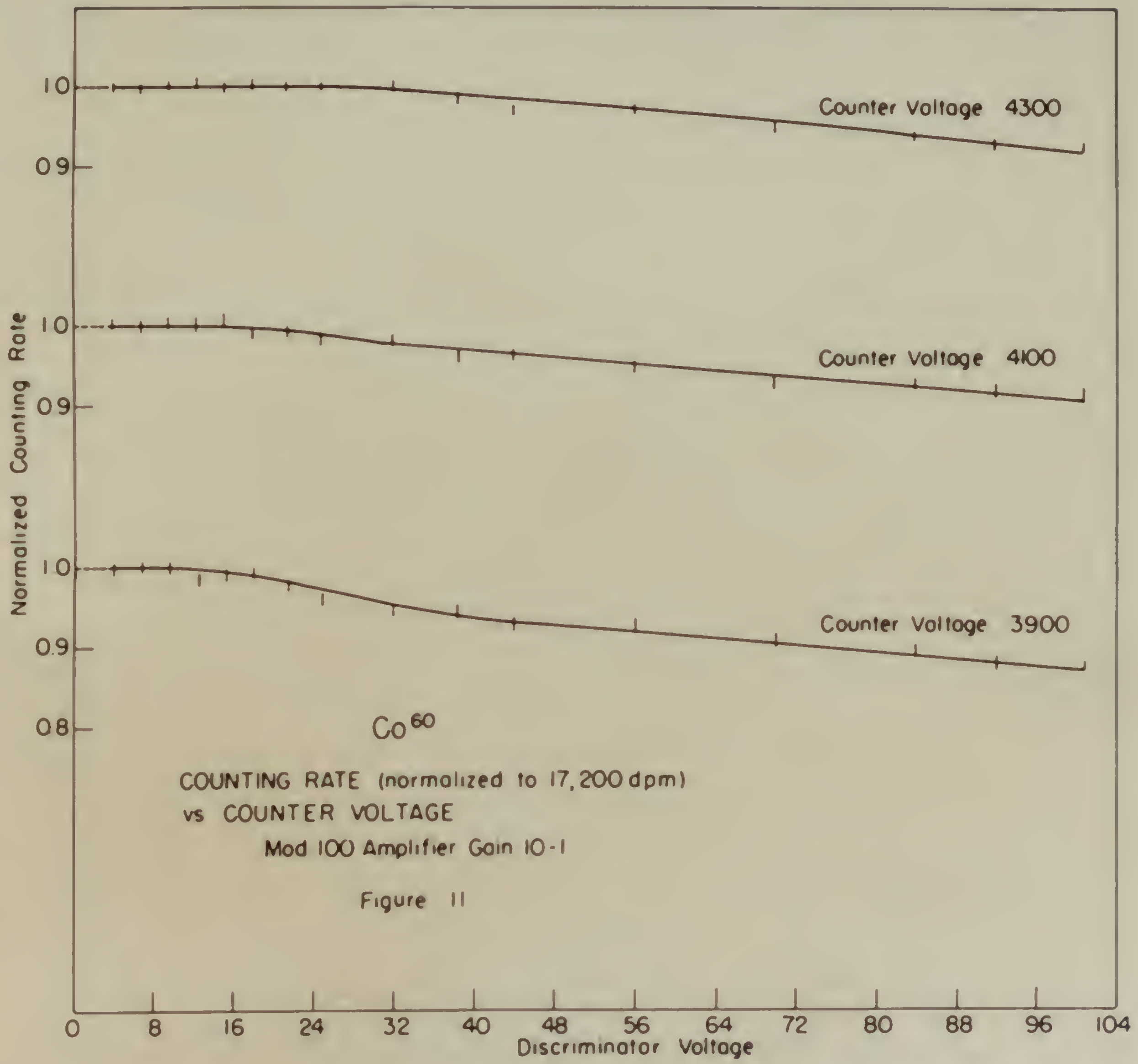


P^{32}

COUNTING RATE (normalized to 38747 dpm)
vs COUNTER VOLTAGE

Mod 100 Amplifier Gain 10-1

Figure 10



Co^{60}

COUNTING RATE (normalized to 17,200 dpm)
vs COUNTER VOLTAGE
Mod 100 Amplifier Gain 10-1

Figure 11

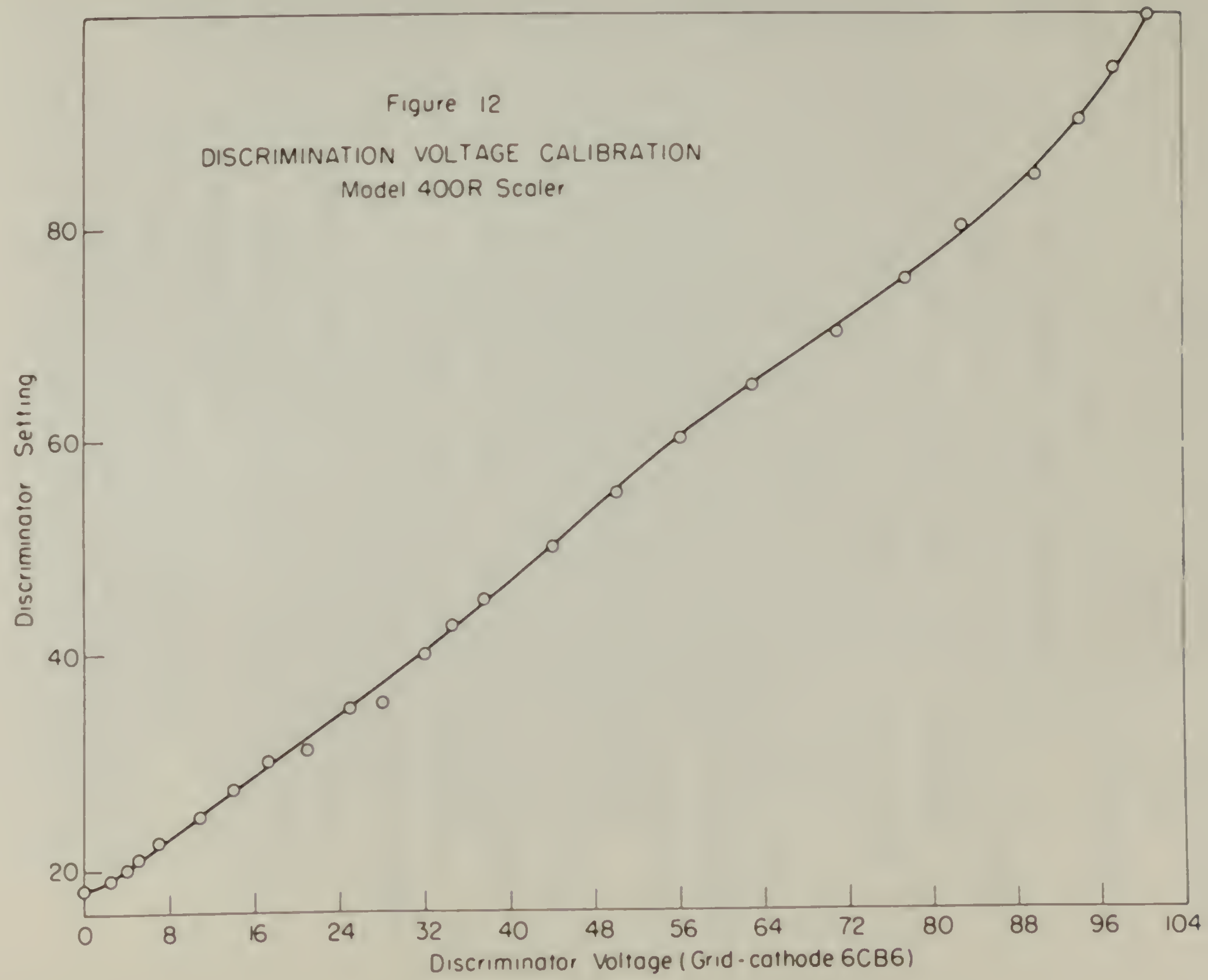


Figure 12

DISCRIMINATION VOLTAGE CALIBRATION
Model 400R Scaler

disintegration rate is indicated on all curves. A further check to verify proper operation is a statistical analysis of the counting rates for points on the discriminator plateau. The mean value thus obtained should agree with the extrapolated true counting rate.

Once the voltage range of the discriminator plateau has been determined for a given isotope the counting procedure is simplified. The discriminator may then be set at the midpoint of the voltage plateau and with counter voltage set at 4300 volts and an electronic gain of 10:1, a series of runs is made. A statistical analysis of these runs is then made to insure that the counter is operating properly and the observed counting rate is determined by the mean value thus obtained.

An electronic gain of 10:1 is chosen as the normal operating point. At this value the largest pulses in the counter just overdrive the Model 100 amplifier without causing counting losses, and the smallest pulses are sufficiently larger than noise to be detectable over a useful discriminator range.

3. Correction to observed counting rate.

The average background is subtracted from the mean value of the measured counting rate, to yield R_0,

the observed counting rate. For counting rates $\leq 60{,}000$ dpm, this value may be used as N_t, the true disintegration rate, if an error of 3 percent is acceptable.

Any or all of the following corrections may be made depending on the degree of precision desired:

a. Correction for resolving time losses. If N_o and N_A are defined respectively as observed and actual disintegration rates, then

$$N_A = \frac{N_o}{1 - N_o t}$$

when t is the counter resolving time which has been determined to be approximately 20 µsec.

b. Correction for absorption due to source mounting film. If it is desired to make this correction, then at the same time the source mountings are prepared, cover layers of parlodion film of the same thickness should be prepared on source rings having a central hole of 1 3/8 inch diameter. The conducting film evaporated on these cover layers should be of the same thickness as that deposited on the ring containing the active source.

With the active source in place obtain N_{A_1}, the actual counting rate. Then remove the source ring,

carefully place the ring containing the cover layer directly over the source to form a sandwich. With this sandwiched source in place, again measure N_{A_2}. The percentage difference in N_{A_1} and N_{A_2} should be quite close to the true correction for absorption due to the mounting film.

A slightly more accurate determination of absorption in the mounting film$^{(7)}$ is quoted below for completeness.

"Experiments have been conducted to determine the amount of absorption, if any, due to the film between the source and the lower half of the 4π counter. The number of particles counted by the top half of the counter connected separately will be

$$N_{top} = \frac{N_o}{2}\left[1 + B_f + (1 - \tau)B_w(b)\right] \qquad (1)$$

where N_o is the true disintegration rate of the source, B_f is the percentage backscattering from the film, is the fractional absorption in the film, and $B_w(b)$ is the percentage backscattering due to the walls in the bottom half. The number of particles counted by the bottom half connected separately will be

$$N_{bottom} = \frac{N_o}{2}\left[(1 - \tau) + (1 + B_f)B_w(t)\right] \qquad (2)$$

"From symmetry considerations $B_W(t) = B_W(b) = B_W$.

"The factor B_F can be neglected when the film is thin and of low atomic number, so that (1) becomes

$$N_{top} = \frac{N_o}{2}(1 + B_W - \tau B_W) \qquad (3)$$

In the bottom half, again assuming $B_F = 0$, one obtains

$$N_{bottom} = \frac{N_o}{2}\left[(1 - \tau) + B_W\right] \qquad (4)$$

Putting this in the form $y = ax + b$ gives

$$N_{bottom} = \frac{-N_o}{2}\tau + \frac{N_o}{2}(1 + B_W). \qquad (5)$$

"With the thin films under consideration it can be assumed that the absorption is directly proportional to the film thickness. Equation (5) can be used to determine the absorption correction graphically. A more direct method of determining τ can be deduced from eq. (3) and (4).

$$N_{top} - N_{bottom} = \frac{N_o}{2}\tau(1 - B_W). \qquad (6)$$

The actual counting rate observed with top and bottom halves connected together is

$$N_{tb} = N_o(1 - \tau/2) \qquad (7)$$

so that (6) becomes, if one lets $N_{top} - N_{bottom} = \Delta$

$$\tau = \frac{N_{top} - N_{bottom}}{N_{tb} - N_{bottom}} = \frac{\Delta}{N_{tb} - N_b}, \qquad (8)$$

and similarly

$$B_W = \frac{N_{top} + N_{bottom}}{N_{tb}} - 1. \qquad (9)$$

The absolute counting rate is then obtained by substituting (8) into

$$N_o = \frac{N_{tb}}{1 - \tau/2}. \qquad (10)$$

"Thus by taking three different readings of the same source on a single film it is possible to determine the absorption by the film. This proves extremely useful for low energy β particles."

c. Corrections for self-absorption and backscattering due to finite source thickness. In general the sources prepared are very thin compared with the half thickness for β absorption in source material. Since the resultant self-absorption and backscattering corrections are small (usually $<$ 1 percent), approximate methods may be used to compute these corrections. The average source thickness is computed from the source area and the known mass of material contained therein.

Self-absorption may be estimated as follows:

Let t = half thickness for β in source material (in $\mu g/cm^2$)

$\bar{x}$ = average source thickness (in $\mu g/cm^2$)

then the true activity N_t is related to the observed activity N_A by

$$N_A \simeq \frac{N_t}{\bar{x}} \int_0^{\bar{x}} \left(\tfrac{1}{2}\right)^{(\bar{x}/t)} d\bar{x}$$

$$\frac{N_A}{N_t} = \frac{t}{\bar{x}} \left[\frac{1 - \left(\frac{1}{2}\right)^{\bar{x}/t}}{\ln 2}\right] = \frac{1 - \left(\frac{1}{2}\right)^{\bar{x}/t}}{0.693\,\frac{\bar{x}}{t}}$$

and $$\frac{N_A}{N_t} \simeq 1 - (0.346)^{\bar{x}/t} \text{ for } \bar{x} \ll t$$

If desired, backscattering corrections may be approximated from the results published by Zumwalt.(9) In these corrections it is assumed that when the source mounting material is very thin, the percentage of saturation backscattering obtained is a function only of its thickness in terms of absorption half thickness. With this assumption, the Zumwalt data obtained for polystyrene can be applied to parlodion by comparing relative half thicknesses involved.

There is also a small loss for particles which travel transversely through the film and are absorbed

before emerging. An order of magnitude approximation of this loss can be made by consideration of the solid angle within which particles will traverse one half-thickness of the film before emerging. For P^{32} (assuming a half thickness of about 100 mg/cm^2) this gives, for a uniform film 0.05 mg/cm^2 thick:

$$\delta\Omega \sim \frac{2\pi \times \frac{0.05}{100}}{4\pi} \simeq \frac{1}{4000}$$

where $\delta\Omega$ = the fraction of emergent particles which traverse a path $\geq$ one half-thickness of the film, which is negligible. For softer β particles, this correction may be large enough to require consideration.

H. Important Characteristics of 4π Counter

Counter voltage plateau: begins at 3700 volts, slope < 0.6 percent per 100 volts.

Settings for normal operation:

Counter voltage: 4300 volts

Cathode: 2500 volts below ground, 600 volts supplied by batteries.

Center wires and guard rings: 1800 volts above ground.

Electronic gain (Model 100 amplifier, Atomic Instrument Co. preamplifier Model 204B):

Course gain: 10
Fine gain: 1

Resolving time: 20 ± 5 μsec (determined by a series of measurements by the two-source method)

n-butane flow gas rate: approximately 2 bubbles/sec

Flushing time required for stable operation: 20 min

Length of discriminator plateau (at normal operating settings):

Co^{60}	25 volts
I^{131}	30 volts
P^{32}	50 volts

Counter efficiency:

$Co^{60} \simeq$ 87 percent (due to high solid content of available Co activity)

$I^{131} \geqslant$ 98 percent

$P^{32} \geqslant$ 99 percent

Appendix I - Bibliography

1. Caswell, R. C.: Ph.D. Thesis, "A Study of the Average Energy of β-Rays," MIT (1947).
2. Borkowski, C. J.: Conference on Absolute β-Ray Counting, National Bureau of Standards, July 13, 1949.
3. Seliger, H.: Private communication, 1953.
4. Rossi, B. B. and H. H. Staub: Ionization Chambers and Counters, McGraw-Hill Book Co., N. Y., 1939.
5. Brown, S. C.: "Theory and Operation of G-M Counter", Nucleonics, 2, 10 (1948); 3, 50(1948); 3, 46 (1948).
6. Seliger, H.: Private communication, 1953.
7. Seliger, H. and L. Cavallo: "The Absolute Standardization of Radioisotopes by 4π Counting", J. Research Natl. Bur. Standards, 1,41 (July 1951).
8. Putnam, J. L.: "On the Absolute Measurement of β-Emitters", United Kingdom Atomic Energy Research Establishment AERE-NR-318, 24 February 1949.
9. Zumwalt, L. R.: United Kingdom Atomic Energy Research Establishment, MDDC-1346. (Reproduction of curves contained in reference no. 8).

10. Houtermans, F. G., L. Meyer-Schutzmeister, and D. H. Vincent: "Energy Calibration for 4π Counters", Z. Physik. **1**, 134 (1952).

APPENDIX II

THE SCINTILLATION γ-RAY SPECTROMETER

APPENDIX II

THE SCINTILLATION γ-RAY SPECTROMETER

The counter consists of a sodium iodide thallium activated crystal 4.3 cm in diameter and 5 cm long mounted on an RCA type 5819 photomultiplier tube. The gain of the linear amplifier used is adjusted so that the spectrum is always represented by voltage pulses of from 0 to 100 volts. A single channel differential pulse height analyzer, fed by the linear amplifier, drives a precision counting rate meter of variable time constant and full-scale sensitivity of from 200 to 20,000 counts per minute. The discriminator base line is varied continuously from approximately 0 to 100 volts and a window of 2 volts is used for all observations. Calibration runs were made before and after obtaining each set of data by use of γ-emitters of known energy.

APPENDIX III

THE END WINDOW β COUNTER

APPENDIX III

Table of Contents

Section A. Description of Equipment 78

Section B. Experimental Technique 78

Section C. Corrections of Observed Data 79

Section D. Resolving Time Losses 80

APPENDIX III

List of Figures

Figure No.	Title	Page
1	End window β counter response curve . .	82
2	Curve of lost counts per minute	83

APPENDIX III

THE END WINDOW β COUNTER

A. Description of Equipment

This equipment, used in determining β energies, consists of a conventional end window Geiger-Muller tube* and a set of calibrated aluminum absorbers.** The tube is mounted in a shielded sample changer*** which contains sliding trays for accurate positioning of source and absorber.

B. Experimental Technique

With a source inserted on the lower tray under the Geiger tube counting rates were recorded for various thicknesses of absorber contained on the upper tray.

---

* Tracerlab, Inc. Model TG-C2 Geiger Tube, window thickness 3 mg/cm^2.

** Tracerlab, Inc. Type R-3A Calibrated Absorbers.

*** Tracerlab, Inc. Model SC-90 Shielded Manual Sample Changer.

Counting procedure varied depending upon the thickness of absorber. At high counting rates, the number of counts per 1 minute interval was recorded for at least three intervals. At intermediate counting rates, the preset count feature of the scaler was utilized and the time required for 10,000 counts was recorded. At very low counting rates, a preset count of 4000 was used.

The tube was operated at the mid-point of its voltage plateau to insure maximum stability. In addition a set of five standardized β emitters of known energy was periodically counted thus enabling the correction of observed counting rates for any changes in instrument sensitivity.

C. Corrections of Observed Data

Other than correcting for counter sensitivity fluctuations mentioned above, the only correction required was for resolving time losses. These corrections were made by adding to the observed counting rates the number of lost counts per minute (Fig. 2).

D. Resolving Time Losses

It is generally assumed that the resolving time of an instrument is constant for all counting rates and corrections are usually made for resolving time losses by means of equations derived on the basis of two general counter types; the paralyzable and the non-paralyzable. A detailed treatment of these two cases* results in the following equations:

$$n = Ne^{-N\rho} \quad \text{(paralyzable type)} \qquad (1)$$

$$n = N(1 - n\rho) \quad \text{(non-paralyzable type)} \qquad (2)$$

At low counting rates both equation (1) and equation (2) reduce to

$$N = n(1 + n\rho) \qquad (3)$$

where N and n are respectively the true and observed counting rates and ρ is the resolving time.

In this experiment it was considered necessary at times to count at very high rates (~ 50,000 cpm) because of the possible presence of short-lived isotopes and the desire to obtain complete sets of absorption data as quickly as possible. It was obvious that the approximate equation (3) could not be used and it was also found that

---

* Evans, R. D.: Class Notes for Course 8.512, Chapter 30.

neither equations (1) nor (2) properly corrected the observed counting rates if a constant resolving time was assumed.

To obtain a useful relation between counting rate and resolving time loss, the response of the instrument to a series of standard sources of known activity was measured and a plot made of observed vs expected counting rate (Fig. 1).* Two response curves are shown, one for a discriminator setting of 4, the other for a setting of 6.** From the curve for a discriminator setting of 4, the setting used throughout the experiment, a plot of lost counts per minute vs observed counting rate (Fig. 2) was prepared to facilitate correction of the observed data.

To verify the accuracy of this procedure, several sources were counted with discriminator settings of 4 and 6. The following tabulation of the counting rates observed and the true counting rates computed from the applicable curve of Fig. 1, shows that the computed values agree within experimental error thus indicating that consistent corrections may be made by this method.

---

* Evans, R. D.: Class Notes for Course 8.512, Chapter 30, page 15.

** Data of E. Samuels, Physics Research Laboratory, Massachusetts General Hospital, Boston, Mass.

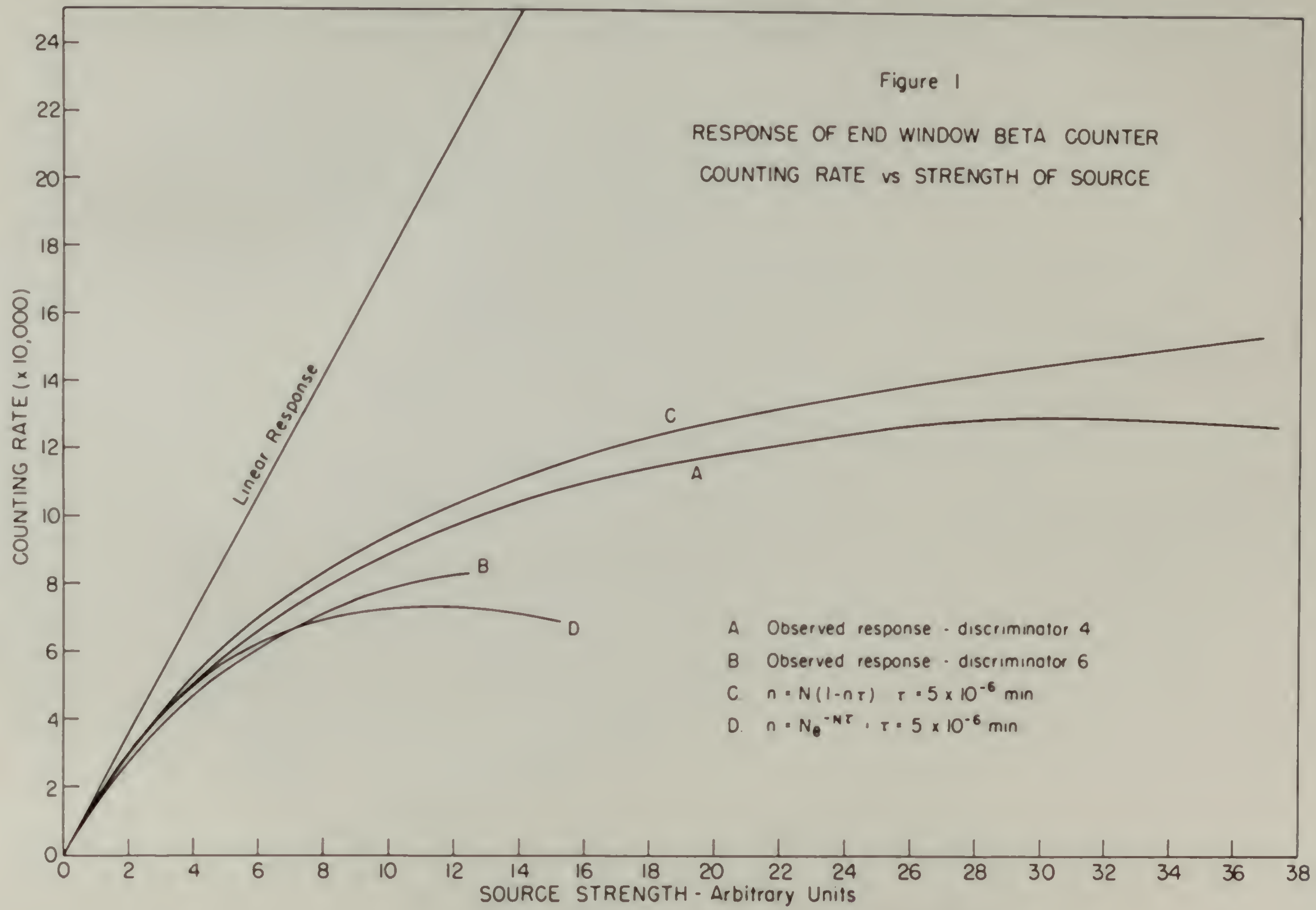


Figure 1

RESPONSE OF END WINDOW BETA COUNTER COUNTING RATE vs STRENGTH OF SOURCE

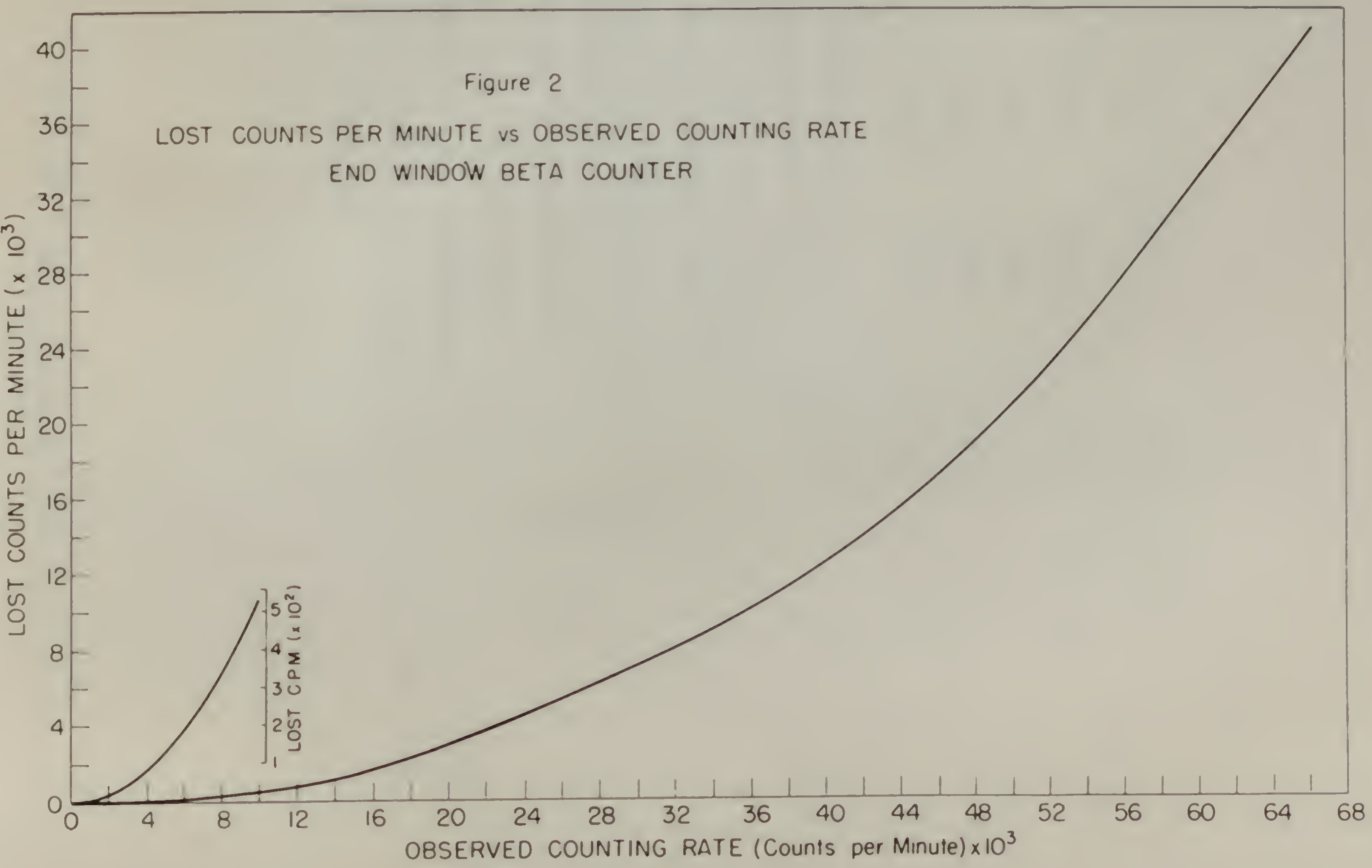


Figure 2

LOST COUNTS PER MINUTE vs OBSERVED COUNTING RATE
END WINDOW BETA COUNTER

Observed counting rate cpm		Computed true counting rate cpm	
Disc. 4	Disc. 6	Disc. 4	Disc. 6
10580	9850	10990	10700
41350	38910	54350	53900
67600	61100	110,000	109,000

Referring to Fig. 1, it is seen that neither response curve coincides with the theoretical curves of equations (1) and (2). It is apparent that the number of lost counts is strongly dependent upon the discrimination level. The results obtained may be explained by a consideration of the pulse height distribution as a function of counting rate.* This shows that as the counting rate is increased, many small pulses are formed and some fraction of these pulses is lost because of the discrimination level and not because of the dead time of the tube.

---

* McCall, R. C.: "Geiger-Muller Counters", M.I.T. Progress Report, 1953.

APPENDIX IV

THE COINCIDENCE COUNTER

APPENDIX IV

Table of Contents

Section A. Description 88

Section B. Experimental Technique 91

Section C. Correction of Observed Counting Rates 96

APPENDIX IV

List of Figures

Figure No.	Title	Page
1	Schematic diagram of scintillation counter	90
2	Coincidence counter assembly	93
3	Sensitivity distribution of coincidence counter	95
4	Coincidence counter response curve . . .	98

APPENDIX IV

THE COINCIDENCE COUNTER

A. Description

This equipment consists of two thallium activated sodium iodide scintillation counters connected in coincidence with single channel and coincidence scaling circuits driving mechanical registers. The crystals are 1.5 inches in diameter, 1 inch deep, and are mounted on RCA type 5819 photomultiplier tubes.

The counters are contained in lead shielded heads along with their cathode follower type preamplifiers shown schematically in Fig. 1. The two heads are mounted on a mechanical scanning device such that the two opposing crystals are coaxial and are separated by approximately 27 cm. A mounting bracket permits positioning of a source equidistant from the crystal faces and colinear with their common axis.

The coincidence circuit is of conventional design* providing both single channel and coincidence outputs which

---

* Dwg. No. B-1547-A, file 6425, Laboratory for Nuclear Science, M.I.T., 28 April 1950.

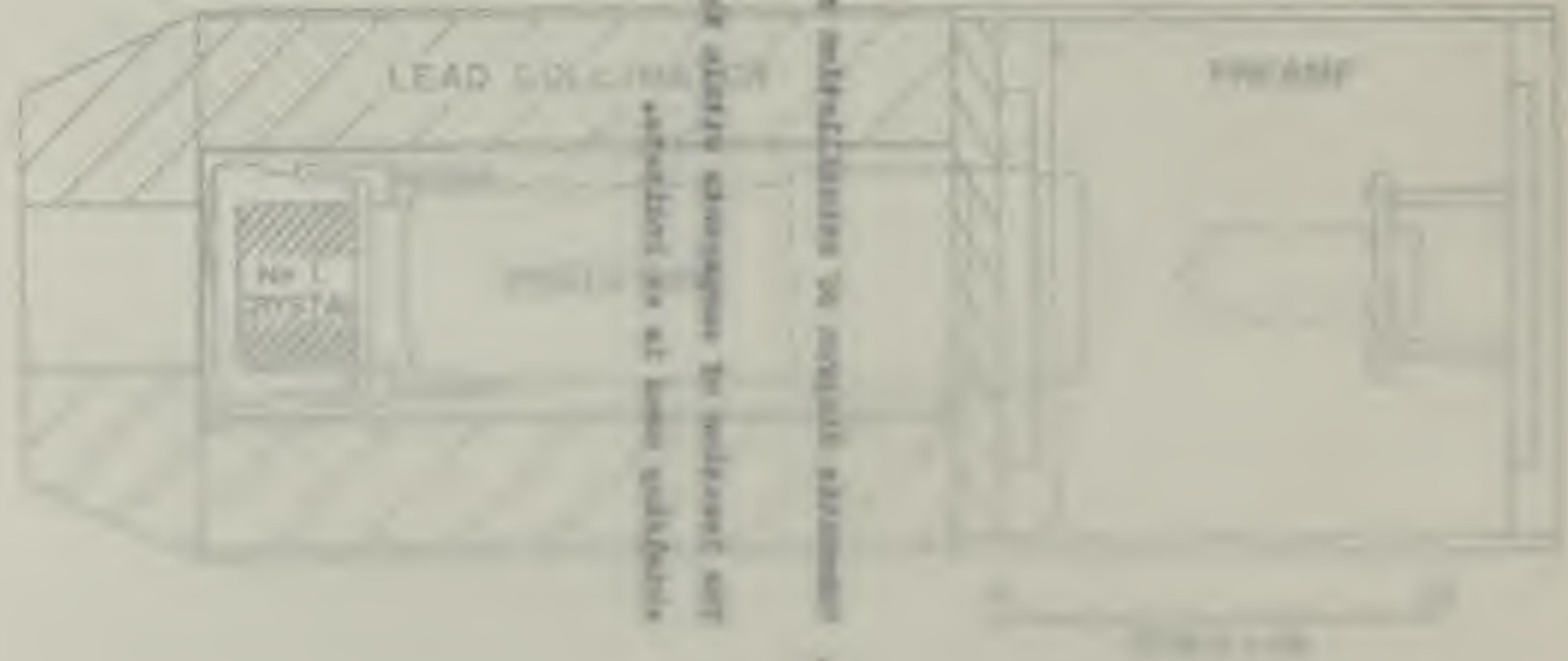
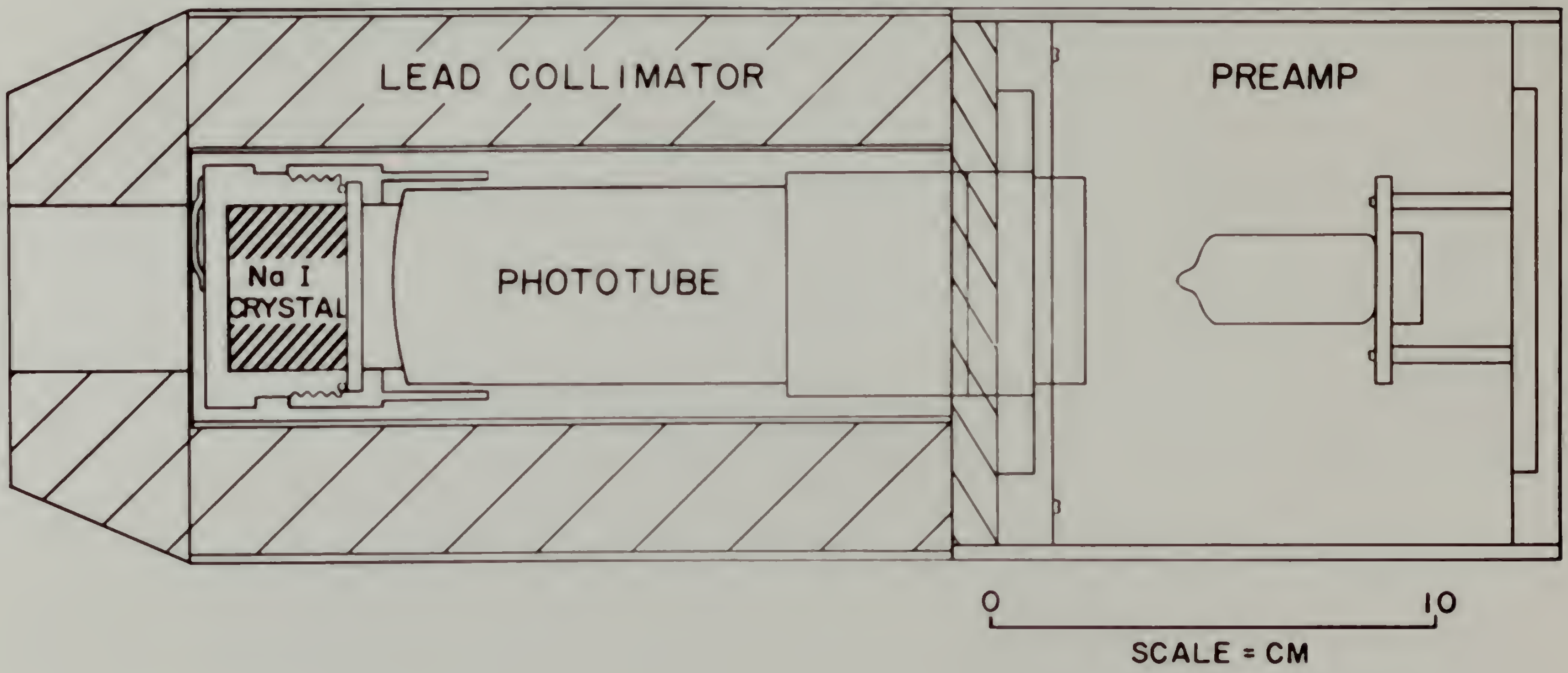

LEAD COLLIMATOR
NaI CRYSTAL
PREAMP

Fig. 1. Schematic diagram of scintillation counter.

The location of components within the lead shielding head is as indicate.


LEAD COLLIMATOR
PREAMP
Na I
CRYSTAL
PHOTOTUBE
0
10
SCALE = CM

are fed through linear amplifiers* to separate scaling circuits**. The equipment assembled for normal use is illustrated in Fig. 2.

B. Experimental Technique

The equipment is operated so that the individual channels register approximately equal counting rates when a source is at the mid-point on the axis between the counter heads.

Figure 3 illustrates that counting rates are only slightly affected by small displacements of the source from its central position. To minimize errors caused by variation in counter sensitivity due to other causes, a standard Na^{22} source was counted prior to each measurement and the correction thus determined was applied to the observed counting rate.

In all measurements the number of counts per 1 minute interval was recorded, each observation including at least three intervals for single channel counts and six intervals for coincidence counts. At least 10,000 events were included in each observation to insure a maximum fractional standard deviation of $\leq$ 1 percent.

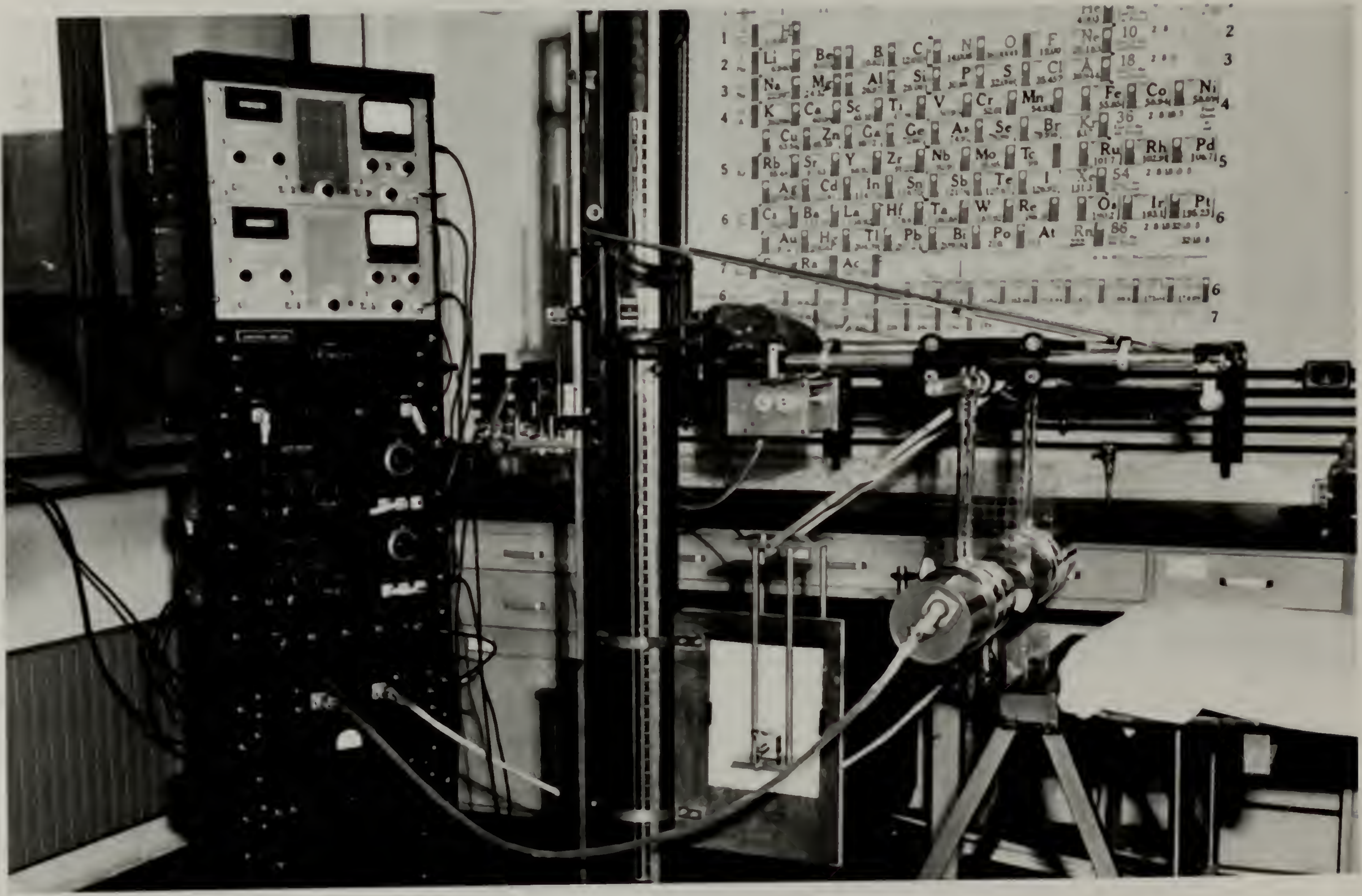
---

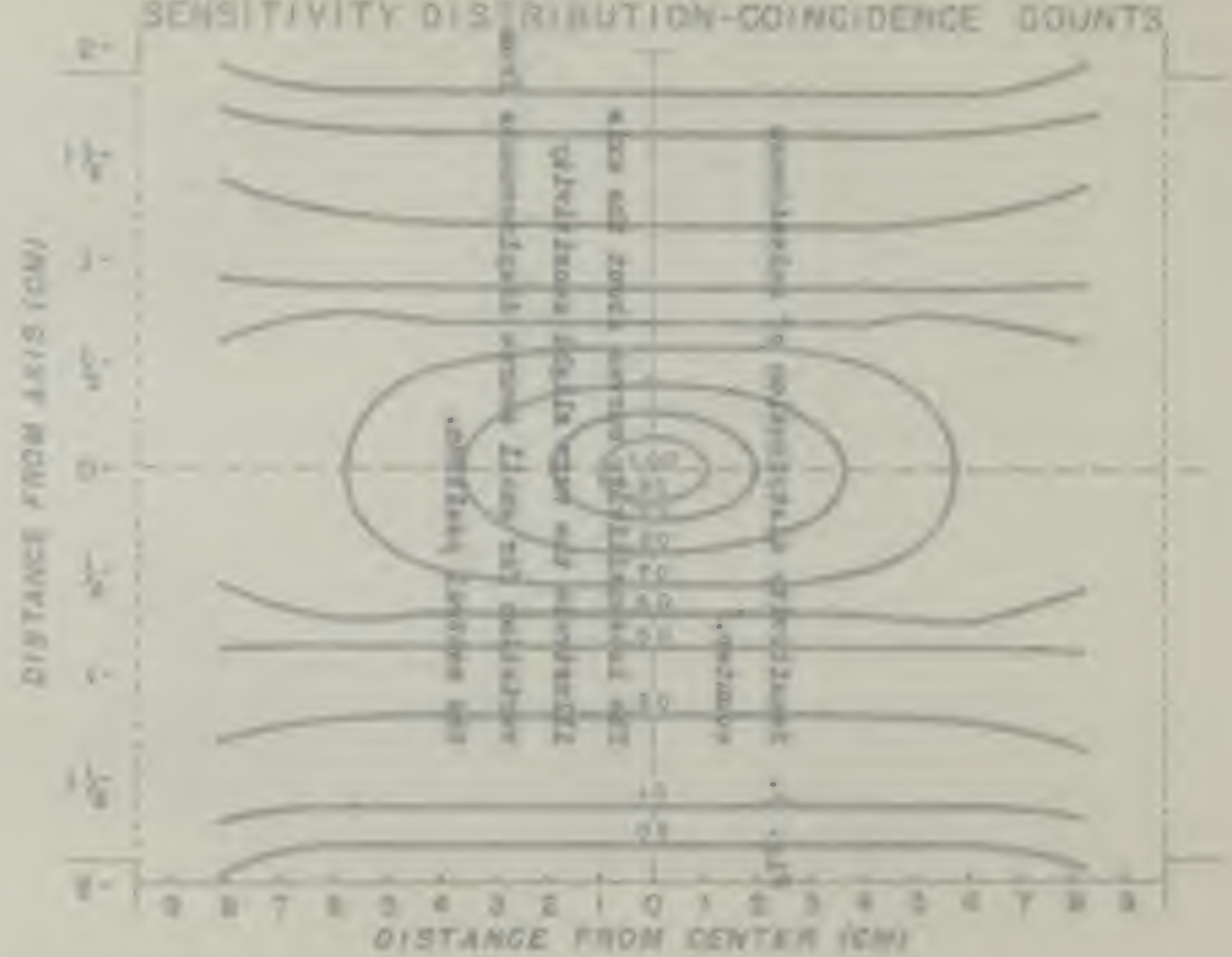
* Atomic Instrument Co. Model 204B Linear Amplifier.

** Atomic Instrument Co. Model 1030 "Scale of 1000" Scaler.

Fig. 2. Coincidence counter assembly.

The equipment is shown as assembled for use in scanning measurements at Massachusetts General Hospital. The scanning and plotting mechanisms are contained in the central section of the photograph. The opposing lead shielded counter heads are visible to the right of the plotting board.



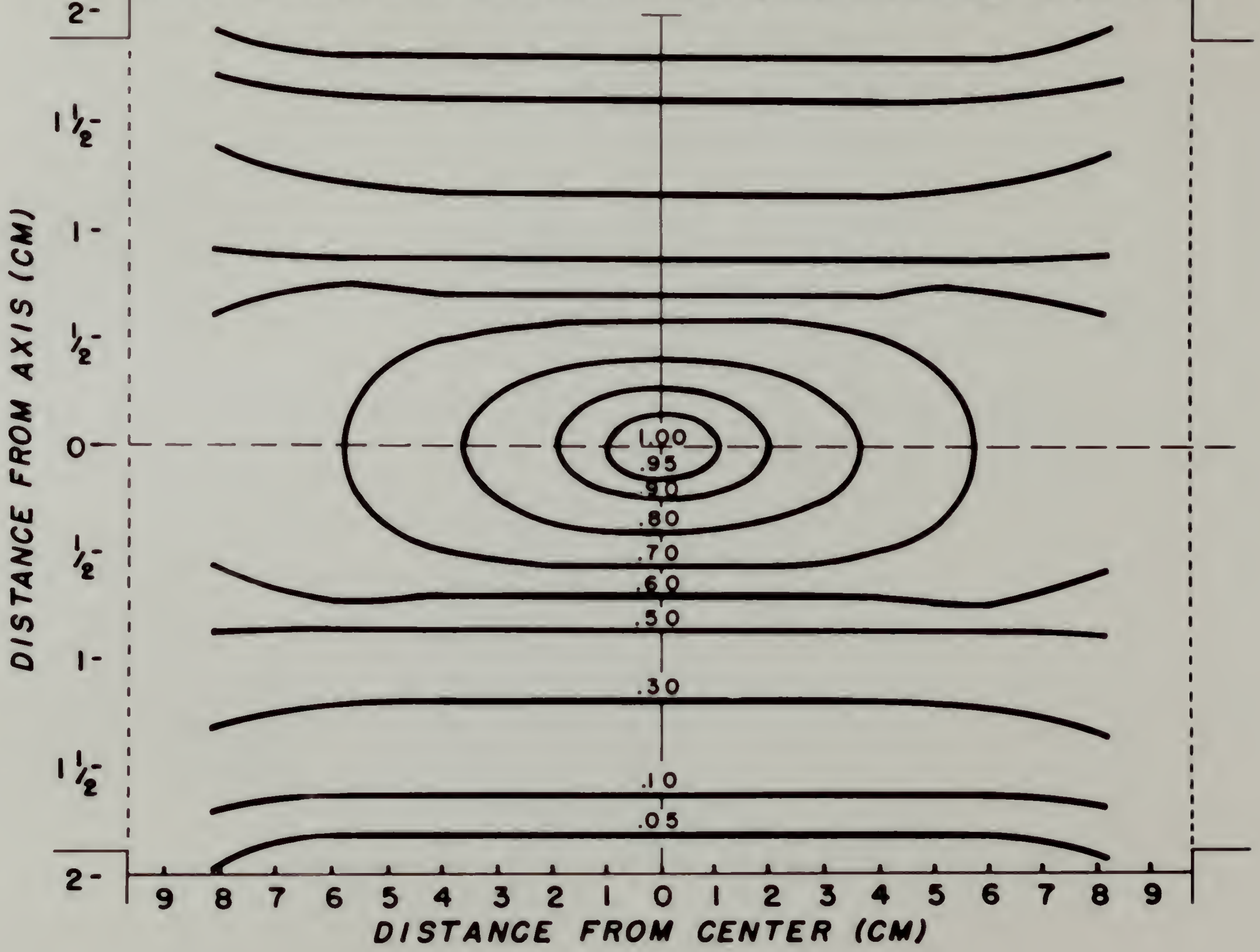


SENSITIVITY DISTRIBUTION-COINCIDENCE COUNTS
DISTANCE FROM AXIS (CM)
2
1½
1
½
0
½
1
1½
2
DISTANCE FROM CENTER (CM)
9
8
7
6
5
4
3
2
1
0
1
2
3
4
5
6
7
8
9
1.00
.80
.70
.60
.50

Fig. 3. Sensitivity distribution of coincidence counter.

The isosensitivity curves about the axis illustrate the very slight sensitivity variation for small source displacements from the central position.

SENSITIVITY DISTRIBUTION-COINCIDENCE COUNTS



C. Correction of Observed Counting Rates

Coincidence counting rates were corrected by subtracting from the observed values the chance coincidence rate. Chance rate was computed by means of the equation

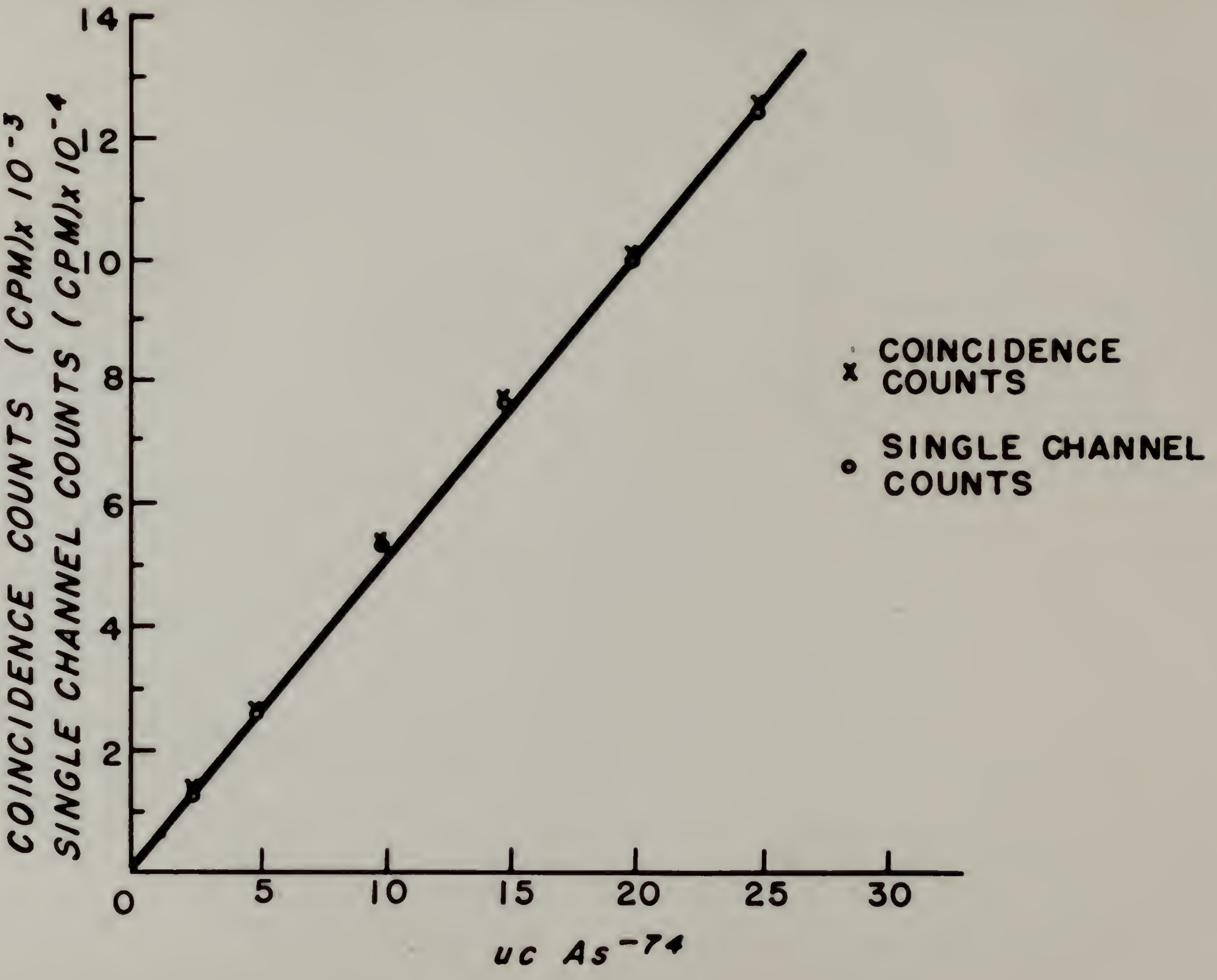
$$C_{ch} = 2\,\tau N_a N_b$$

where N_a, N_b are the individual channel rates and τ is the resolving time of the coincidence circuit. By counting an essentially monoenergetic γ-ray emitter positioned off the axis of the crystals, τ was computed to be approximately 0.36 μsec by use of the above equation.

Counter response appears to be linear for counting rates up to 140,000 cpm on single channels and 14,000 cpm for coincidences (Fig. 4). Consequently no corrections were applied to the data for resolving time losses.

Fig. 4. Coincidence counter response curve.

The linearity of counting rate vs source strength is evident for both single channel and coincidence counts.


COINCIDENCE COUNTS (CPM) x 10^-3
SINGLE CHANNEL COUNTS (CPM) x 10^-4
14
12
10
8
6
4
2
0
5
10
15
20
25
30
uc As-74
x COINCIDENCE COUNTS
o SINGLE CHANNEL COUNTS

JUL 2
APR 22
BINDERY
461

20653

Thesis W27

Watters

An investigation of the radionuclides of arsenic produced by cyclotron bombardment of germanium with 15 Mev deuterons.

JUL
BINDERY
★
APR 22
BINDERY
461

20653

Thesis W27

Watters
An investigation of the radionuclides of arsenic produced by cyclotron bombardment of germanium with 15 Mev deuterons.

Library
U. S. Naval Postgraduate School
Monterey, California

ARTISAN
GOLD LETTERING
AND
SMITH BINDERY
OAKLAND, CALIF.

thesW27
An investigation of the radionuclides of
3 2768 001 93021 7
DUDLEY KNOX LIBRARY